## **FINAL REPORT**

# Technical Summary Report for the Historical Public Exposures Studies for Rocky Flats Phase II

Part of Task 6: Technical Support for Public Involvement

September 1999

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### **CONTENTS**

INTRODUCTION	<u>1</u>
Historical Public Exposures Studies on Rocky Flats	2
Plant History and Description	_
Estimating Risks from Releases of Radionuclides and Chemicals to the Environ	
Review of the Historical Records	
	_
MATERIALS AND EXPOSURE PATHWAYS	<u>5</u>
Plutonium	<u>6</u>
Carbon Tetrachloride	<u>7</u>
Other Contaminants Examined during Phase II	<u>7</u>
Tritium	
Beryllium	<u>8</u>
Dioxins	<u>9</u>
Uranium	<u>10</u>
MODELING APPROACH	<u>11</u>
Identifying Discrete and Continuous Releases	<u>11</u>
Model Domain and Receptor Grid	
Atmospheric Dispersion and Transport	<u>12</u>
Exposure Scenarios	<u>13</u>
Risk Evaluation	<u>14</u>
Plutonium	<u>14</u>
Carbon Tetrachloride	<u>15</u>
Beryllium	<u>16</u>
Uncertainty	<u>17</u>
ATMOSPHERIC RELEASES AND RESULTING RISKS	<u>18</u>
Releases of Plutonium	<u>19</u>
Routine Operations	<u>19</u>
1957 Fire	<u>22</u>
1969 Fire	<u>24</u>
903 Area	<u>26</u>
Summary of Plutonium Releases	<u>29</u>
Risks from Plutonium Releases	<u>29</u>
Releases of Carbon Tetrachloride	<u>33</u>
Releases of Beryllium	<u>34</u>
ESTABLISHING CONFIDENCE IN THE RESULTS	36
Comparisons with Soil Measurements	
Comparisons with Air Measurements	
Comparisons with Measurements in Other Media	
PUBLIC INTERACTION AND RISK COMMUNICATION	<u>45</u>

CONCLUSIONS AND RECOMMENDATIONS	<u>47</u>
REFERENCES	<u>50</u>
APPENDIX A: PHASE I AND PHASE II REPORTS	.A-1

### **FIGURES**

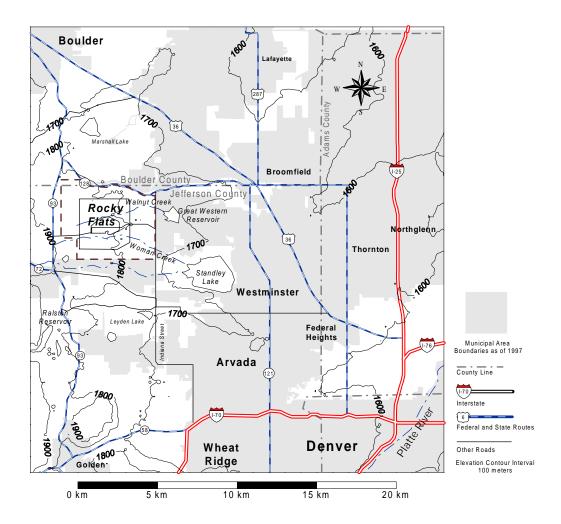
1.	Location and key environmental features of the Rocky Flats Environmental	
	Technology Site	<u>1</u>
2.	Model domain and receptor grid for the Phase II study	<u>13</u>
3.	Atmospheric release points modeled for plutonium, beryllium, and	
	carbon tetrachloride	<u>18</u>
4.	Estimates of routine plutonium releases (µCi)	20
5.	Estimated annual plutonium concentrations in air east of the RFP on Indiana Street	
	outside the current buffer zone	<u>21</u>
6.	Estimated 9-hour average plutonium concentration in air 1 m (3.3 ft) above ground	
	at the 50 <sup>th</sup> percentile level during the 1957 fire	<u>24</u>
7.	Ground-level time-integrated concentration of plutonium at the 50 <sup>th</sup> percentile level	
	for respirable particles (<15 µm AED) released from the 903 Area between 1964 to 1969	<u>28</u>
8.	Lifetime cancer incidence risk from plutonium inhalation for the laborer scenario:	
	5 <sup>th</sup> percentile and 95 <sup>th</sup> percentile	<u>30</u>
9.	Map showing assumed locations for the laborer scenarios used to calculate cancer risk	<u>31</u>
10.	Lifetime cancer incidence risk from plutonium inhalation for the laborer scenario at	
	selected locations in the model domain	<u>32</u>
11.	Lifetime cancer incidence risk from carbon tetrachloride inhalation for the	
	laborer scenario: 5 <sup>th</sup> percentile and 95 <sup>th</sup> percentile	<u>34</u>
12.	Annual release estimates for beryllium with uncertainty as estimated by	
	<u>ChemRisk</u> (1994b)	35
13.	Estimated and measured plutonium soil concentrations in the 0–3-cm (0–1.2-in.)	
	layer for three transects originating from the 903 Area and extending eastward	<u>39</u>
14.	Comparison of plutonium soil inventory estimates from soil concentration	
	measurements	<u>41</u>
15.	Comparison of estimated annual average plutonium concentrations in ambient air with	
	measurements at four locations in the model domain	<u>43</u>
16.	Annual average plutonium concentrations in ambient air from 1953 to 1989 at	
	Indiana Street and plutonium-specific measurements taken from 1970 to 1989	<u>44</u>

## **TABLES**

1.	Dose conversion factors (rad μCi <sup>-1</sup> ) for inhalation of plutonium oxide aerosols	<u>14</u>
2.	Lifetime cancer incidence risks per unit dose (rad <sup>-1</sup> )	<u>16</u>
	Estimated plutonium releases during the 1957 fire	
4.	Estimated plutonium releases during the 1969 fire	25
5.	903 Area discrete release estimates for plutonium attached to	
	<30 μm AED soil particles	<u>27</u>
6.	903 Area continuous release estimates (1964–1969) for plutonium attached to	
	<30 μm AED soil particles	<u>27</u>
7.	Plutonium release estimate distributions by event	
8.	Lifetime cancer incidence risk from plutonium inhalation for the laborer	
	scenario at selected receptor locations in the model domain	<u>33</u>
9.	Carbon tetrachloride source term estimates	33

#### INTRODUCTION

The Rocky Flats Environmental Technology Site is owned by the U.S. Department of Energy (DOE) and is currently operated by the Kaiser-Hill Company. For most of its history, the site was called the Rocky Flats Plant (RFP) and was operated by Dow Chemical Company as a nuclear weapons production, research, and development facility. The RFP is located on 2650 ha (6500 acres) of Federal property about 8–10 km (5–6 mi) from the cities of Arvada, Westminster, and Broomfield, Colorado, and about 26 km (16 mi) northwest of downtown Denver. Since 1975 the original 156-ha (385-acre) main production area has been surrounded by a large buffer zone that now defines the RFP boundary (see Figure 1).



**Figure 1.** Location and key environmental features of the Rocky Flats Environmental Technology Site. The municipal boundaries illustrated are accurate as of 1997. Urban areas were much smaller during most of Rocky Flats Plant operations (1953–1989).

#### **Historical Public Exposures Studies on Rocky Flats**

Through a 1989 Agreement in Principle between DOE and the State of Colorado, DOE provided the State with funding for health-related studies that were to be directed by the Colorado Department of Public Health and Environment (CDPHE). The purpose of the Historical Public Exposures Studies on Rocky Flats is to evaluate the doses and potential health impacts to nearby residents who may have been exposed to past contaminant releases. This health study focused on members of the public living offsite. A separate joint CDPHE study conducted by the University of Colorado Health Sciences Center is addressing worker exposures. The CDPHE first invited a national panel of experts to help design the health studies. A Health Advisory Panel (HAP) was established with the responsibility of overseeing the health studies. This panel decided to stress public involvement and to separate the research into two major phases conducted by two different contractors to enhance accountability and credibility.

Phase I of this study was carried out by ChemRiska. In Phase I, ChemRisk conducted an extensive investigation of past operations and releases from the RFP. Phase I identified the primary materials of concern; determined release points, events, quantities released, and transport pathways; and made preliminary estimates of offsite dose and risk. The conclusions from Phase I were released in a public summary document (HAP 1993) and a series of task reports by ChemRisk.

Phase II of the study was performed by *Radiological Assessments Corporation*<sup>b</sup> (*RAC*) and involved an in-depth investigation of the potential doses and risks to the public from the key historical releases from the RFP. One key question of interest was whether the public doses and risks warranted an epidemiological study. Phase II was divided into the six tasks listed below.

- **Task 1** Coordinate with ChemRisk to ensure quick and efficient access to the records and individuals contacted by ChemRisk during Phase I of the project
- **Task 2** Verify the radionuclide and chemical release estimates and associated uncertainties that were developed during Phase I of the project
- **Task 3** Conduct an independent assessment of the risks from past Rocky Flats operations using state-of-the-art methods to ensure those risks to the public are carefully identified
- **Task 4** Evaluate historical environmental data, which can provide a basis for risk assessment, reconstructing releases, and validating model estimations
- **Task 5** Provide recommendations for additional offsite measurements to ensure that new measurements focus on the most important locations and releases
- **Task 6** Provide support for the public involvement efforts.

This report summarizes our methods and findings reported during Phase II of the project. More detailed results of the project can be found in the reports listed in <u>Appendix A</u> of this technical summary report.

<sup>&</sup>lt;sup>a</sup> ChemRisk<sup>®</sup>, A Service of McLaren/Hart Environmental Services, formerly A Division of McLaren/Hart, Alameda, California.

b In 1998 Radiological Assessments Corporation changed its name to Risk Assessment Corporation. For consistency throughout the project, all reports were published by Radiological Assessments Corporation.

#### **Plant History and Description**

Construction of the RFP began in 1952, and the first nuclear weapons components produced at the plant were shipped offsite in 1953. Rocky Flats has been a government-owned, contractor-operated facility, and it was originally run by the Atomic Energy Commission. In 1974, the U. S. Energy Research and Development Administration succeeded the Atomic Energy Commission. The U.S. Energy Research and Development Administration was in turn succeeded by the DOE in 1977. The Dow Chemical Company was the original contractor for operations at the RFP. Rockwell International replaced Dow in 1975 and operated the RFP through 1989. EG&G Rocky Flats, Inc. took over from Rockwell in 1990. In 1994, the Kaiser-Hill Company became the plant operator, succeeding EG&G, and it remains the current contractor for the site. Nuclear operations were conducted from 1953 to 1989.

The primary mission of the RFP was to produce components for nuclear weapons from materials including plutonium, uranium, beryllium, and stainless steel. Additional plant missions have involved plutonium reprocessing and waste management. Production activities included metal fabrication and assembly and chemical recovery and purification of transuranic radionuclides. During plant operations, materials were released to the environment either through routine operations or accidentally. These releases went to air, surface water, and indirectly to groundwater. Some contaminants have been detected in groundwater under the site, but the contamination has not yet traveled offsite. Because the Historical Public Exposures Studies on Rocky Flats address exposures and risks to the public living offsite, the groundwater pathway was not considered in Phase II. It is emphasized that the groundwater pathway may be an important potential pathway of exposure to the public in the future, and it is being considered in other studies.

In 1989, Rocky Flats was placed on the Superfund National Priorities List. In 1991, the DOE entered into a cleanup agreement with the U.S. Environmental Protection Agency (EPA) and the CDPHE. Production ceased in 1992, and current activities at the site center on environmental cleanup and restoration, waste management, and consolidation of materials.

<u>Figure 1</u> shows the general location and key environmental features of the site. Approximately 2.5 million people currently live within an 80-km (50-mi) radius of the site. Adjacent land use is a mixture of agriculture, open space, industry, and low-density housing. The area is rapidly growing, especially in the corridor between Boulder and Denver and to the west in the direction of Rocky Flats.

The climate is characterized by dry, cool winters and warm summers. Winds, though variable, come predominately from the northwest. The site's sloping geographical location and its proximity to a major mountain range can create dramatic changes in temperature and rapidly changing weather conditions. Of particular importance in our research were the strong westerly winds that develop during the passage of synoptic weather fronts and that often occur in winter and early spring. This environmental factor was particularly important because of concerns about resuspension of dust containing plutonium from soil around the site and the potential exposures to persons living downwind. Annual precipitation averages slightly greater than 38 cm (15 in.), with more than 80% of the precipitation occurring between April and September. The elevation of the site is approximately 1860 m (6100 ft) on the eastern edge. To the east, the topography slopes gradually at an average downgrade of 29 m (95 ft) per mile. Surface drainage at the site occurs in a west to east pattern along several ephemeral streams. Historically, North Walnut Creek, South

Walnut Creek, and Woman Creek drained from the main plant facilities area into the Great Western Reservoir and Standley Lake. The reservoir and lake were used as municipal water supplies until the 1990s when diversion dams and stream flow modifications ensured that neither received surface water from the RFP. The transport of materials from the site through surface water pathways was also of concern to the public and was evaluated in Phase II.

Understanding the impact of past releases from the Rocky Flats Environmental Technology Site is extremely important because of its proximity to such a highly populated area, its popularity because of its agreeable climate and proximity to the Rocky Mountains, and potential future expansion of habited areas near the site.

#### Estimating Risks from Releases of Radionuclides and Chemicals to the Environment

The process of estimating risks to people from radionuclides and chemicals released to the environment can best be described using the following illustrative equation:

$$Risk = (S \times T \times E \times D \times R)_{uvcp}$$
(1)

where

S = source term (quantity and type of material released)

T = environmental transport and fate of the material released

E = exposure factors (characteristics of individuals exposed that affect their intakes)

D = conversion of intakes to dose

R = conversion of dose to risk

u = uncertainty analysis

v = validation

c =communication of results

p = public participation.

Each of the areas listed above could be considered a discipline in itself, but the combination of these different areas of expertise gives us estimates of risk for an exposed population. The source term characterizes the amount, type, and length of time the materials were released to the environment. In Phase II, we performed a detailed analysis of specific sources of plutonium, carbon tetrachloride, and several other materials of concern that have been released from Rocky Flats since the beginning of site operations. We studied the transport of these materials through the environment using environmental data and mathematical models. Exposure scenarios that describe the lifestyle, residence location, and duration of exposure were used to characterize contaminant intake via inhalation for hypothetical individuals in the model domain. The exposure scenarios were designed to help people understand whether they were exposed and the amount of exposure they may have received. We used organ-specific plutonium risk coefficients developed specifically for this project and slope factors published by the EPA for chemicals to convert contaminant intake to estimates of incremental lifetime cancer incidence risk.

Finally, the estimates of risk must consider uncertainties, validation, communication of the results, and public involvement in the process. Parameter uncertainty analysis helps us understand the precision of our model estimates. Validation compares the estimated concentrations in

environmental media to corresponding measured data that were collected over the years. Validation is a measure of model accuracy and is one means by which we assess the uncertainty in the model formulation. Risk communication is the process of documenting the results so people can understand them. The public must be involved throughout the process for credibility and to consider their concerns and ideas. This report summarizes how Phase II of the study addressed each of these areas and the results of our study. Complete documentation of Phases I and II of the study is available in a comprehensive set of reports listed in Appendix A.

#### **Review of the Historical Records**

Researchers involved in both Phases I and II conducted extensive searches of the historical records to find information to support the Historical Public Exposures Studies on Rocky Flats. Although most of the records were located onsite or in the Denver area, some records were found in storage areas at other DOE sites and at Federal repositories.

In some cases, information that was needed was classified and had to be declassified before it could be released. Throughout the project, we made every attempt to make all records used in the project available to the public to ensure independent verification of the data used. All information requested for our analysis was made available; no information was withheld because of classification or privacy issues. Over the course of the project, we used thousands of records to help reconstruct releases to the environment, to make estimates of risk, and to help verify our calculations. Although the document search was the most extensive ever conducted on Rocky Flats, as with all historical dose reconstruction efforts, many records could not be found. These records had either been destroyed as part of the routine destruction of records over the years, or they were simply not located in spite of extensive search. Nevertheless, based on the evidence from the historical records, we believe our research has identified the major sources of releases to the environment and exposure to people.

#### MATERIALS AND EXPOSURE PATHWAYS

More than 8000 materials were used or stored at the RFP. Some of these were part of the production processes, but most were typical chemicals found in industrial facilities. These materials were released as by-products of the routine operations at the site. A careful evaluation initially done in Phase I, and confirmed during Phase II, indicated that of the more than 8000 materials, plutonium<sup>c</sup> and carbon tetrachloride were the most important contaminants released into the environment in terms of potential exposure and associated risk to the public. Plutonium was processed and fabricated into weapons components at the RFP throughout the period of operation. As a result, it was released to the air and water in greater quantities than most of the other radioactive materials used at the site. Carbon tetrachloride is a solvent that was used to clean plutonium processing machinery and instruments. A number of other contaminants were also identified for further investigation in Phase II, primarily because members of the public

<sup>&</sup>lt;sup>c</sup> In this context plutonium means weapons grade plutonium, which consists primarily of <sup>239</sup>Pu (~93.8%), <sup>240</sup>Pu (5.8%), and <sup>241</sup>Pu (~0.36%) by weight percent. Specific activity of weapons grade plutonium is 0.072 Ci g<sup>-1</sup>.

expressed concern that these materials received inadequate treatment in Phase I. These additional contaminants were tritium, beryllium, dioxins, and uranium.

The following sections provide information about the major sources and uses of these contaminants at the RFP. The sections discuss criteria used to identify the relevant exposure pathways for each contaminant and to determine if more detailed risk calculations for contaminants other than plutonium and carbon tetrachloride were warranted.

#### Plutonium

The principal plutonium isotopes of concern at the RFP are <sup>239</sup>Pu and <sup>240</sup>Pu, which have long half-lives of 24,065 years and 6,537 years, respectively. Plutonium emits alpha particles that are the least penetrating type of radiation, but they create short, dense ionization trails. Alpha particles have such weak penetration abilities that they can be blocked by a piece of paper or the outer layers of the skin. As a result, the prime danger from an alpha-emitting radionuclide like plutonium comes from having it inside the body. For residents in the vicinity of Rocky Flats, plutonium is most likely to have entered the body from breathing air that contained plutonium particles released from the site. Plutonium was released to the atmosphere during routine industrial production from 1953 to 1989 and as a result of two major fires in plutonium processing buildings in 1957 and 1969. In addition, plutonium-contaminated soil from a barrel storage area, called the 903 Area, was blown offsite during windstorms, mainly during 1968 and 1969. These atmospheric plutonium releases were the focus of much of Phase II and are described later in this report.

The Phase I results (<u>ChemRisk</u> 1994a) showed soil ingestion and inhalation of resuspended soil-bound plutonium to be minor pathways when considering the long-term exposure to atmospheric releases from Rocky Flats. Although these pathways become increasingly important for the later years of exposure because of the accumulation of deposited plutonium in soil and the lower airborne emissions directly from the RFP, the doses during 1971–1989 were several orders of magnitude smaller than doses for earlier years (1952–1970).

Plutonium and other radioactive contaminants, such as tritium, were also released into creeks near the RFP, which flowed offsite into drinking water reservoirs east of the plant. Waterborne wastes were treated and released into holding ponds; then water from the ponds was released gradually. Releases to water entered one of the two major creeks that drained the RFP: Walnut Creek (which has north and south branches onsite) or Woman Creek. The creeks also collected storm runoff from the plant site, which contained numerous waste storage burial and incineration areas near the plant's central industrial area. Walnut Creek, which drains about 50% of the RFP site, flows into Great Western Reservoir in northeastern Jefferson County about 2.4 km (1.5 mi) from the plant. In 1989, the City of Broomfield built a diversion ditch around the reservoir to prevent Rocky Flats surface drainage and runoff from entering the reservoir via Walnut Creek. In 1997, the City of Broomfield secured a new drinking water supply; and the Great Western Reservoir is no longer used as a drinking water source. Woman Creek drains about 35% of the RFP site, particularly the southeastern part. Until recently, Woman Creek flowed from the RFP site into Standley Lake, a drinking water supply for the Cities of Westminster, Thornton, and Northglenn and some residents of Federal Heights. In 1996, the new Woman Creek Reservoir was completed to prevent Rocky Flats surface water from flowing into Standley Lake via Woman Creek. Water in the new reservoir is then pumped to Walnut Creek below the Great Western Reservoir, where it flows downstream into Big Dry Creek and the South Platte River.

During Phase I of the project, water monitoring data for radioactivity in drinking water supplies from reservoirs downstream of the RFP were analyzed for their potential impact on offsite populations (ChemRisk 1994b). Conservative screening calculations were used to calculate radiation doses associated with the elevation of radioactivity measured in the Great Western Reservoir, which has been linked to Rocky Flats releases. The results suggested that some contamination of the primary reservoirs from Rocky Flats releases was possible. In Phase II, we compared the estimated exposures from the surface water pathway with historical measurements taken in the environment. This comparison required some knowledge of the methods used onsite to collect, transport, and treat liquid wastes before they were discharged offsite. Thus, our focus was on the liquid waste handling system at the RFP before the mid-1970s.

#### **Carbon Tetrachloride**

Large quantities of carbon tetrachloride (CCl<sub>4</sub>), a highly volatile solvent, were used at Rocky Flats to clean and degrease product components and equipment. Rocky Flats was formerly the largest volume consumer of carbon tetrachloride in the United States. Most of the carbon tetrachloride evaporated into the air and was released in building ventilation exhausts.

Inhalation of carbon tetrachloride is the exposure pathway of greatest concern for releases from Rocky Flats. Carbon tetrachloride does not bioconcentrate and <a href="ChemRisk">ChemRisk</a>® (1994a) estimated the dose from direct inhalation is at least 1800 times higher than indirect exposure pathways that involve transfer from air through animals and plants before transfer to humans. No monitoring for carbon tetrachloride released to surface water or soil was conducted. Carbon tetrachloride rapidly volatilizes from soil and surface water and would have likely evaporated from creeks and reservoirs within days of its release. Therefore, concentrations in these media were expected to be inconsequential compared to concentrations in the ambient air.

We evaluated the small amount of air monitoring data that were found, reviewed the evidence of health effects, and estimated airborne concentrations and lifetime carcinogenic incidence risks resulting from inhalation of carbon tetrachloride for generic receptor scenarios. It is important to understand that, as is often the case, monitoring data for carbon tetrachloride and other chemicals are not as available as are data for radionuclides. Because of this, estimating risks from exposure to chemicals is often more difficult and uncertain.

#### Other Contaminants Examined during Phase II

#### **Tritium**

Tritium is a radioactive isotope of hydrogen with a 12.5-year half-life that emits low energy beta particles when it decays. Tritium replaces hydrogen in a water molecule so tritiated water is the most common form of tritium in the environment. It reaches surface water (a) directly from precipitation via molecular exchange with water in the atmosphere and direct scavenging of tritium by rain or snow and (b) by direct releases of tritiated water, for example, from the RFP to Walnut and Woman Creeks. Tritium was not produced at the RFP, but it has been released accidentally from the RFP on several occasions during processing of tritium-contaminated scrap

plutonium from Lawrence Livermore Laboratory. On a few occasions, tritium was released unknowingly from the site. In April 1969, 57 Ci was released; in March 1971, 40 Ci was released; in November 1971, 29 Ci was released; in April 1973, 350–1600 Ci was released to air and 50–100 was released to the Great Western Reservoir; and in September 1974, 1.5 Ci was released to air (Meyer and Till 1999).

The tritium release of most concern to the public was the April 1973 incident that released tritium to Walnut Creek, which feeds into the Great Western Reservoir. The main reason for the concern was that the tritium releases to surface water were much higher than any of the other tritium release incidents. Furthermore, the site was not monitoring for tritium at that time because tritium was not a contaminant released during normal operations onsite. Fortunately, the Colorado Department of Health (now called the CDPHE) was monitoring at the site boundary, so tritium measurement data were available to document this accidental release of tritium.

The 1973 tritium release incident occurred when tritium-contaminated scrap material was processed. The incoming material had been checked only for alpha contamination but not for tritium, which is a beta emitter. The processing operations were done at the RFP from April 9–25, 1973. Tritium concentrations did increase in water from the Great Western Reservoir, but there was no measurable difference in tritium concentrations in Standley Lake water before and after the tritium event. This observation is understandable because the plutonium processing areas discharge liquid effluent to the Walnut Creek drainage (see the discussion of liquid effluent treatment in the "Plutonium" section). It was estimated that about 50–100 Ci accumulated in the Great Western Reservoir.

We used the measured tritium concentrations in the Great Western Reservoir following the tritium release event to estimate the radiation dose to an adult from ingestion of Broomfield water during the period of higher-than-normal tritium concentrations. Based on these calculations, the median dose estimate was 0.32 mrem, with  $5^{th}$  and  $95^{th}$  percentiles of 0.16 and 0.58, respectively. For comparison, the committed dose equivalent from natural background levels of tritium in the environment is 0.0012 mrem  $y^{-1}$ . Natural background from all radionuclides in the body amounts to 39 mrem  $y^{-1}$  (NCRP 1987, Table 2.4). Because of the relatively small risks associated with exposure to tritium in the environs of the RFP compared to those associated with other contaminants, detailed source terms and risk estimates were not completed during this phase of the project.

#### **Beryllium**

Beryllium is a nonradioactive metal that was used to make nuclear weapons components. At Rocky Flats, beryllium dust was produced during its machining. The beryllium dust particles were released through vents and stacks at the plant, and in most cases were subjected to the same high-efficiency particulate air (HEPA) filtration as the routine operational emissions of plutonium. Beryllium was released to the air from routine operations and three accidental fires at the RFP from 1958 to 1989.

Beryllium was transferred offsite in the air and in creeks that flowed to surface water used for drinking. Starting in 1980, beryllium was monitored in water effluent. Routine surface water monitoring showed less than the analytical detection limit of 0.05 mg of beryllium per liter of water. Historically, inhalation of beryllium has been a much greater human health concern than ingestion, in part because less than 1% of ingested beryllium is absorbed through the

gastrointestinal tract (<u>EPA</u> 1991). Beryllium is not known to bioaccumulate. In Phase II, we did not evaluate releases of beryllium to surface water further because of a lack of effluent and environmental monitoring data to quantify the releases, insufficient evidence of accumulation in soils and sediments, and low solubility and gastrointestinal absorption.

We reviewed and evaluated several studies of beryllium in soil at the RFP. Available data suggest that atmospheric releases of beryllium from RFP releases are not detectable in soil. We evaluated risk estimates associated with atmospheric releases of beryllium (McGavran and Rood 1999a), and they are described <u>later in this report</u>.

#### **Dioxins**

Dioxins are chlorinated compounds that cause cancer, immunological, developmental, and reproductive effects in experimental animals. Dioxins are released from all combustion processes. Emissions of dioxins from incinerators vary widely depending on the nature of the waste, type of incinerator, operating efficiency, temperature, and air pollution control devices.

A multichambered retort incinerator operated in Building 771 from late 1957 to 1989. The incinerator burned plutonium-contaminated materials such as plastics, paper, rubber, and cloth. The offgas was subjected to scrubbers and filters; it then went to the main building plenum, through a HEPA filter, and was released to the atmosphere from a 43-m (140-ft) stack. After 1968, the offgas was subjected to six banks of HEPA filtration. Ash from the incinerator was sent to a disposal facility or sometimes processed for plutonium recovery. Seven other incinerators or open burning pits were also operated at the RFP at various times.

The incinerator in Building 771 was likely to have been the largest source of dioxins from the RFP because it operated for 32 years and burned chlorinated waste and plastics. Most dioxins released from an incinerator are associated with particulates; therefore, HEPA filters are considered very effective in removing them from the offgas effluent. However, a former RFP incinerator operator indicated that the HEPA filters for the incinerator were often overloaded and were not cleaned or replaced as often as ideal. To help assess the significance of dioxin releases, we performed preliminary screening calculations, also called bounding calculations, to determine potential release, transport, exposure, and cancer risk. Because no monitoring for dioxins was done at any of the facilities, we calculated emissions based on what was known about the throughput and operating parameters for the incinerator.

The HAP held a workshop in February 1995 to talk about methods that might be used to better estimate the formation of dioxins, their release, bioaccumulation up the food chain, exposure, and health risk. A wide variety of opinions existed on the appropriate parameter values. To obtain a range of values, we asked various experts in the field of dioxin emissions from incinerators for their opinion of the best values to use for these parameters. The risk estimates for dioxin releases developed from this workshop are believed to be upper bounds rather than best estimates for two main reasons: 1) the cancer potency factors used were developed by the EPA and correspond to the 95<sup>th</sup> percentile risk values and 2) reduction of dioxins in airborne effluent from HEPA filtration was not considered.

The central value for the distribution of median risk estimates for cumulative dioxin releases from 1953–1989 was  $3 \times 10^{-9}$ . The 5<sup>th</sup> and 95<sup>th</sup> percentiles of the distribution were  $5 \times 10^{-10}$  and  $1 \times 10^{-7}$ , respectively. These risk estimates are likely to overestimate the true risk. The HEPA filters used for the incinerator likely reduced dioxin emissions considerably. The conservative

risk estimates we calculated for the RFP are less than the risk range of  $10^{-6}$  to  $10^{-4}$  that is used by the EPA to regulate carcinogenic pollutants.

As a follow up to these screening calculations, we explored several activities that might have provided information about the magnitude of released dioxins. We investigated the practicability of analyzing ash from the incinerator in Building 771, which is still stored at the RFP, to find out if dioxins were formed in measurable quantities in the incinerator. Because the amount of dioxins in the bottom ash might not correlate to the amount released and considering the expense and difficulties associated with analyzing ash containing plutonium, the HAP decided not to pursue ash sample analysis. We investigated the feasibility of analyzing dioxins in blood lipids of incinerator workers or nearby residents. Many difficulties, including the expense, the time that had passed since the incinerator operated, and questions about the ability to detect differences in the concentrations of dioxins in people to be tested, led us to recommend and the HAP to decide that study on human subjects was not justified.

In response to questions raised by members of the public, we also collected information on other waste incinerators operating in the Denver area over the past 40 years. These included municipal waste, hazardous waste, and medical waste incinerators. The Rocky Mountain Arsenal's Submerged Quench Incinerator was the largest incinerator to operate in the Denver area. It operated from June 1993 through July 1995 and burned about 50 times the maximum bounding value of  $3.9 \times 10^5$  kg y<sup>-1</sup> throughput used for the screening assessment emissions estimates for dioxins released from the incinerator in Building 771. In light of the large releases elsewhere and because of the significant uncertainty associated with estimating dioxin releases from the RFP, further detailed investigations into releases of dioxin were not conducted.

#### Uranium

Uranium is a naturally occurring radioactive metal that, like plutonium, emits radioactivity in the form of alpha particles. Enriched uranium was used at the RFP to make nuclear weapon parts. Voillequé (1999a) documents screening calculations to assess the radiological significance of atmospheric releases of uranium from the RFP relative to plutonium. We evaluated routine atmospheric releases of uranium from buildings, open outdoor burning of depleted uranium metal chip waste, and a conservative assessment of an accidental outdoor fire involving depleted uranium. Based on the Phase II evaluation of routine plutonium releases (Voillequé 1999b), we applied two adjustments to the Phase I routine uranium release estimates (ChemRisk 1994b) to account for nonrepresentative sampling of the uranium concentration in the large exhaust ducts and to account for the effects of self-absorption. The largest amounts of uranium were released in the mid-1950s. We also evaluated releases to surface water and the chemical toxicity of uranium.

Routine releases of plutonium were 4.5 times more significant radiologically than enriched uranium<sup>d</sup> and 2.2 times more significant radiologically than depleted uranium. Furthermore, routine operational releases of plutonium were small relative to the major accidental releases (particularly the 903 Area and the 1957 fire), further diminishing the overall importance of uranium. A conservative assessment of the accidental burning of a pallet of depleted uranium concluded that the radiological significance of uranium released was 60 times less than one high

<sup>&</sup>lt;sup>d</sup> Enriched uranium has a higher than natural content of the fissionable isotope <sup>235</sup>U, and depleted uranium has less. Because enriched uranium was more valuable, it was controlled more carefully and releases were smaller than for depleted uranium.

wind day of releases from the 903 Area (<u>Voillequé</u> 1999a). These results substantiate the emphasis on plutonium as the primary radionuclide with respect to historical public exposures.

#### MODELING APPROACH

This section describes the mathematical modeling approach we used for estimating the lifetime cancer risk resulting from plutonium, carbon tetrachloride, and beryllium releases from the RFP. Mathematical models help translate release estimates from the site into contaminant concentrations in the environment where people live, using available site-specific data. Several elements of the approach are critical to understanding the dose and risk calculations that emerge from this modeling effort. These elements include the types of events for which dose and risk were calculated, the model domain or geographical area of concern that was defined for this project, the atmospheric transport calculations, the historical exposure scenarios, and the types of uncertainty associated with the calculations.

The conceptual model describes the four steps involved in the Phase II calculations: source term, atmospheric transport modeling, exposure scenarios, and risk evaluation. Source term defines the quantity and timing of the contaminants released to the atmosphere. We coupled the source term to an atmospheric transport model to calculate contaminant concentrations in air within the model domain. We assumed various exposure scenarios to calculate contaminant intake via inhalation to hypothetical individuals residing in the model domain. We then converted contaminant intake to organ-specific incremental lifetime cancer incidence risk, using risk coefficients for plutonium and slope factors for nonradiological contaminants.

#### **Identifying Discrete and Continuous Releases**

An important distinction for Phase II of this project was the type of release for which source term, exposure, and risk was explored. For the most part, the releases that emerged from Phase I as important to further investigate were releases that occurred over relatively short periods of time ( $\leq$ 26 hours). In this project, these releases are referred to as discrete releases.

Discrete releases at the RFP were the result of two glove box fires that occurred in 1957 and 1969 that resulted in airborne releases of plutonium, and a series of wind-driven suspension events where plutonium-contaminated soil was suspended from the 903 Area and transported downwind. Therefore, we reviewed three discrete releases for this work: the 1957 fire in Building 771, the 1969 fire in Buildings 776/777, and releases from the 903 Area largely during high wind events that occurred on 6 days in 1968 and 1969.

Continuous releases occurred over the operational history of the facility. In particular, routine operational releases of plutonium occurred from the Building 771 stack and roof vents for Buildings 776/777 and other buildings. We also treated the suspension of plutonium-contaminated soil from the 903 Area during typical meteorological conditions from 1964 to 1969 as a continuous release. Releases of nonradiological contaminants (particularly carbon tetrachloride and beryllium) were also approximately continuous.

Understanding the breakdown of releases into discrete and continuous events is central to understanding the estimated risks from exposure. For discrete events, exposure was dependent on spatial location and timing. If an individual was not in the path of the contaminant plume at the time of the event, that individual was not exposed to the release. In contrast, for continuous

events, exposure occurred throughout the model domain, and the magnitude of exposure was mainly a function of direction and distance from the plant.

#### **Model Domain and Receptor Grid**

We developed a domain early in the project within which contaminant concentrations, receptor exposure, and cancer risk were calculated. The model domain (Figure 2) encompassed a 2200 km² (860 mi²) area that extended 28 km (17.5 mi) south, 12 km (7.5 mi) west, 22 km (13.75 mi) north, and 32 km (20 mi) east from center of the RFP. Most of the Denver metropolitan area and the city of Boulder are included in the domain. The domain was limited in its western extent because few individuals live there and most of the contaminants traveled east and southeast of the plant. The open circles in Figure 2 define grid nodes, or locations at which calculations within the model were made and individuals could be located.

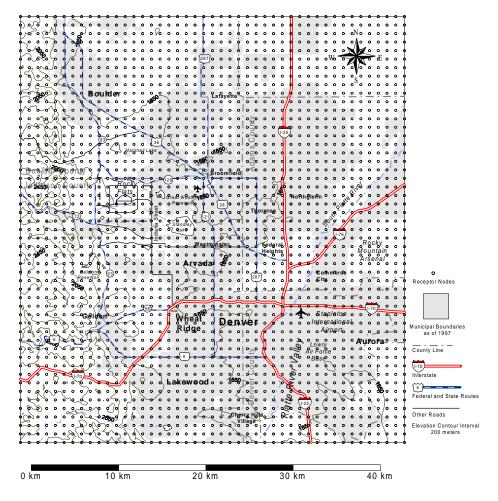
#### **Atmospheric Dispersion and Transport**

Atmospheric transport modeling was performed using the RATCHET model (Ramsdell et al. 1994). We selected RATCHET based on the results of a model comparison study involving five models (which ranged in complexity from a simple straight-line Gaussian Plume model to a complex terrain model [Rood 1999a]) and results of a site-specific tracer study conducted in 1991. While no model consistently outperformed the others, estimated concentrations from RATCHET exhibited better correlation to observed values overall. In addition, RATCHET incorporates spatially varying meteorological and environmental parameters and modules that perform random sampling of the meteorological parameters, which allowed for a Monte Carlo analysis of uncertainty.

Atmospheric transport simulations were performed differently for discrete and continuous events. For discrete events (Rood and Grogan 1999a, 1999b, 1999c), meteorological data for the specific days of the event were available. We ran RATCHET using the Monte Carlo sampling features that sample from distributions of the basic transport parameters for each Monte Carlo trial.

Continuous events were modeled somewhat differently because meteorological data from the RFP for most of the assessment period were not available. Therefore, we used compilations of recently acquired meteorological data as a surrogate for approximating past conditions. We employed this technique using a 5-year meteorological data set from 1989 to 1993 to estimate annual average plutonium concentrations from routine operational releases and continuous 903 Area suspension releases (Rood 1999b; Rood and Grogan 1999a). We also used this technique to estimate annual average concentrations of carbon tetrachloride and beryllium (McGavran and Rood 1999a, 1999b).

Air concentrations from discrete and continuous releases were integrated over the relevant years of RFP operations at each computational node in the model domain. We combined the time-integrated concentration values with scenario exposure information and risk coefficients to calculate the incremental lifetime cancer incidence risk to hypothetical individuals in the model domain.



**Figure 2.** Model domain and receptor grid for the Phase II study. Receptor nodes are indicated by open circles and define computational nodes at which model calculations were made and individuals could be assumed to be located. Municipal boundaries illustrated are accurate as of 1997. Urban areas were much smaller during most Rocky Flats Plant operations (1953–1989).

#### **Exposure Scenarios**

A key component of the Rocky Flats historical public exposures work was estimating the health impacts to hypothetical individuals in the model domain. The calculated cancer risk to a person from exposure to the contaminants released depended upon a number of factors, such as

- Where the person lived and worked in relation to the RFP
- When and how long that person lived near the RFP (for example, during the key plutonium releases in 1957 and the late 1960s, as opposed to the 1970s when all types of releases were smaller)
- The age and gender of the person
- Lifestyle (for example, did the person spend a great deal of time doing strenuous work outdoors).

Although it is not possible to calculate individual risks for every person who lived or worked in the Rocky Flats area during its operational history, it is also not credible to calculate a single risk value that applies to all residents. To consider the many factors that influence exposure, we developed exposure scenarios of hypothetical residents for whom representative risk estimates could be made. Each scenario represents one individual. These individuals incorporate typical lifestyles, ages, genders, and times spent in the area. The scenarios can help individuals estimate risk ranges for themselves by finding a profile that most closely matches their background. The scenarios were not designed to include all conceivable lifestyles of residents who lived in this region during the time of RFP operations. Rather, they provide a range of potential profiles. Scenarios defined for use in this project included a laborer, an office worker, a homemaker, an infant-child, and a student. The infant-child scenario represented a single individual who matures during the exposure period.

The scenarios were location independent, which allowed us to calculate cancer risk for each scenario at every node in the model domain. In this way, the spatial dependency of risk for each scenario could be observed within the model domain. We reported estimated cancer risks for exposure to carbon tetrachloride, beryllium, and plutonium separately.

#### **Risk Evaluation**

Combining the exposure scenarios with the time-integrated air concentration at the location of interest allowed us to estimate the total contaminant intake via inhalation. We used plutonium risk coefficients developed specifically for this project and slope factors published by the EPA for chemicals to convert contaminant intake to estimates of incremental lifetime cancer incidence risk.

#### **Plutonium**

For plutonium releases at the RFP, inhalation was identified as the primary exposure pathway, and lung, liver, bone, and bone marrow were shown to be the principal organs of concern (<u>Grogan et al.</u> 1999). The dose per unit activity inhaled varied for these four tissues (Table 1). Furthermore, the dose per unit activity (dose conversion factor) also varied depending on the particle size distribution of the inhaled plutonium aerosol (Table 1). We used three different particle size distributions (1, 5, 10-µm activity median aerodynamic diameter [AMAD] and geometric standard deviation of 2.5) to characterize plutonium releases from the RFP.

Table 1. Dose Conversion Factors (rad  $\mu \text{Ci}^{-1}$ ) for Inhalation of Plutonium Oxide Aerosols<sup>a</sup>

Cancer site	Dose conversion factor <sup>b</sup>		
	$AMAD = 1 \mu m^c$	$AMAD = 5 \mu m^c$	$AMAD = 10 \mu m^c$
Lung	$1.2 \times 10^{-8} \ (1.9)$	$7.0 \times 10^{-9} \ (2.7)$	$3.2 \times 10^{-9}$ (4.3)
Liver	$5.4 \times 10^{-9}$ (3.0)	$2.6 \times 10^{-9} $ (3.5)	$1.1 \times 10^{-9}$ (4.5)
Bone	$2.4 \times 10^{-8}$ (3.0)	$1.2 \times 10^{-8} $ (3.5)	$5.7 \times 10^{-8}$ (4.5)
Bone marrow	$1.2 \times 10^{-9}$ (3.0)	$5.9 \times 10^{-10}  (3.5)$	$3.0 \times 10^{-10} \ (4.5)$

<sup>&</sup>lt;sup>a</sup> Reported in units of μGy Bq<sup>-1</sup> in <u>Grogan et al.</u> (1999).

<sup>&</sup>lt;sup>b</sup> Geometric mean is listed with geometric standard deviation in parentheses.

<sup>&</sup>lt;sup>c</sup> Geometric standard deviation of each particle size distribution is 2.5.

Lifetime cancer incidence risk coefficients with uncertainties for plutonium inhalation were developed for lung, liver, bone, and bone marrow (leukemia) as part of the project (<u>Grogan et al.</u> 1999) using four independent sources of information:

- 1. Epidemiological studies of workers exposed to plutonium in Russia
- 2. The dose response relationship observed in the Japanese atomic bomb survivors exposed primarily to gamma (low-linear energy transfer) radiation combined with a relative biological effectiveness factor to account for the difference in biological effectiveness of alpha radiation compared to gamma radiation
- 3. Human dose-response relationships determined for populations exposed to other alphaemitting radionuclides (mainly radon, thorium, and radium)
- The results of controlled experiments with animals exposed to plutonium and other alphaemitting radionuclides.

We combined these independent cancer risk coefficient distributions to develop overall cancer incidence risk coefficient distributions (Table 2) that accounted for both the uncertainties associated with the estimate and an assigned intrinsic merit of the approach. The four primary cancer sites account for ~97% of the total lifetime cancer incidence risk from plutonium inhalation. The influence of gender was accounted for in the analyses (see Grogan et al. [1999] for details). The data also allowed a distinction to be made between the risks and uncertainties to those under 20 years of age at exposure compared to those 20 years old and older. The risk coefficients for persons under 20 years of age were applied to the infant-child and student in the hypothetical exposure scenarios.

#### Carbon Tetrachloride

For carbon tetrachloride, inhalation was the exposure pathway of greatest concern for releases from the RFP. Although acute and chronic exposure to carbon tetrachloride may cause toxic injury to the liver, kidney, and nervous system, liver cancer is the health effect expected to occur at lower exposure concentrations. Carbon tetrachloride is classified as a probable human carcinogen by the EPA (EPA 1998a) because it has been shown to cause liver cancer in several animal species. However, studies of workers exposed to high levels of carbon tetrachloride have not been conclusive.

The excess lifetime cancer risk was determined from the product of the carbon tetrachloride inhalation dose and the cancer slope factor. The EPA cancer slope factor represents the 95% upper confidence limit of the probability of a carcinogenic response per daily unit intake of a chemical over 70 years. We used cancer slope factors established by the EPA for calculating liver cancer risk from carbon tetrachloride inhalation (see McGavran and Rood [1999b] for details). In our analysis, we represented the carbon tetrachloride cancer slope factor by a lognormal distribution having a geometric mean of  $2.5 \times 10^{-2}$  kg d mg<sup>-1</sup> and a geometric standard deviation of 1.4.

		Lifetime incidence risk	
	_	distribution	percentiles <sup>b</sup>
Cancer site	Gender	Age under 20	Age 20 and over
Lung c	Males/ females	13 (1.4 - 90)	13 (1.4 - 86)
Liver	Males	12 (1.5 - 150)	6.3 (0.81 - 80)
	Females	5.7 (0.60 - 80)	3.0 (0.32 - 41)
Bone	Males	0.52 (0.011 - 29)	0.27 (0.0056 - 15)
	Females	0.25 (0.0052 - 14)	0.13 (0.0026 - 7.4)
Bone marrow	Males/females	1.7 (0.041 - 9.3)	1.7 (0.041 - 8.7)

Table 2. Lifetime Cancer Incidence Risks per Unit Dose (rad-1) a

#### Beryllium

Inhalation was the exposure pathway of greatest concern for beryllium releases from the RFP. Beryllium is classified by the EPA as a B1 probable human carcinogen (EPA 1998a) because even though numerous studies have shown that beryllium compounds are carcinogenic in experimental animals by several routes of exposure (including inhalation), there has been debate as to whether beryllium can cause cancer in humans. A number of epidemiological studies have reported an increased risk of lung cancer in beryllium workers, but deficiencies in the studies have not allowed an unequivocal conclusion to be reached.

We used cancer slope factors established by the EPA to calculate lung cancer risk from beryllium inhalation. We assigned a distribution to cancer slope factor values based on the relative risk estimates from an occupational epidemiological study used by the EPA to determine the slope factors (see McGavran and Rood [1999a] for details). In our analysis, we represented the cancer slope factor for beryllium by a triangular distribution having a most likely value of 8.4 kg d mg<sup>-1</sup>, a minimum value of 0.56 kg d mg<sup>-1</sup>, and a maximum value of 25 kg d mg<sup>-1</sup>.

Beryllium can also cause a serious progressive granulomatous disease called chronic beryllium disease. Although the lung is primarily involved, it is a systemic disease and granulomatous inflammation may involve other organs. No clear dose-response relationship or duration of exposure-response relationship has been established for chronic beryllium disease, which is interpreted as involving a delayed hypersensitivity that may be induced by very low exposures. We compared estimated beryllium air concentrations to the EPA's recently established reference concentration (RfC) for beryllium (EPA 1998b). The EPA defines the RfC as an estimate (with uncertainty spanning perhaps an order of magnitude) of a daily exposure to the human population (including sensitive subgroups) that is likely to be without appreciable risk of deleterious effects during a lifetime. The RfC was established based on evidence of sensitization to beryllium that can be detected by measuring in vitro proliferative responses of lymphocytes to beryllium.

<sup>&</sup>lt;sup>a</sup> Reported in units of 10<sup>-2</sup> Gy<sup>-1</sup> in Grogan et al. (1999).

<sup>&</sup>lt;sup>b</sup> 50<sup>th</sup> percentile with 2.5 and 97.5 percentiles in parentheses.

<sup>&</sup>lt;sup>c</sup> Smoking was not accounted for because of lack of information.

#### Uncertainty

An important aspect of the modeling effort undertaken for the RFP was the ability of the model to estimate uncertainty in calculated cancer risks. Environmental assessment models are inherently uncertain. Uncertainty in a model arises because of (a) inexact approximations by the mathematical formulations of physical processes that govern the behavior of the system (model uncertainty) and (b) inexact approximations of model inputs (parameter uncertainty). Model uncertainty may be evaluated by comparing model estimations to measured values that are independent of the data used to develop the model. This process is often termed model validation and is a measure of the model's accuracy. Parameter uncertainty arises because of lack of knowledge about a parameter's true value. The uncertainty in a parameter can result from imprecise measurements or because the parameter represents a quantity that is simply impossible to measure directly and, therefore, must be estimated. A parameter uncertainty analysis provides a measure of the model's precision.

The uncertainty analysis included evaluating both parameter and model uncertainties. We evaluated parameter uncertainty using Monte Carlo simulation combined with simple random sampling to generate an empirical approximation of the probability distribution of the model output. We evaluated model uncertainty through model validation. Model validation involved comparing estimated concentrations in environmental media (air, soil, vegetation, and lake sediment) to corresponding, independently measured data.

We handled parameter uncertainty in the air dispersion process differently for discrete and continuous events. For the discrete events, uncertainty was represented by providing probability distributions of the basic transport parameters (wind speed, wind direction, mixing height, precipitation, and Monin-Obukhov scaling length). For the continuous events, uncertainty was represented using several multiplicative correction factors that accounted for uncertainty in the dispersion process, meteorology, deposition, and plume depletion. Source term and risk coefficient uncertainties were also included in both discrete and continuous release simulations.

Uncertainty in the plutonium risk coefficients, expressed as risk per unit activity intake for a given particle size distribution, accounted for dosimetric uncertainties as well as the uncertainties associated with the approaches used to derive risk per unit dose (see Tables  $\underline{1}$  and  $\underline{2}$ ).

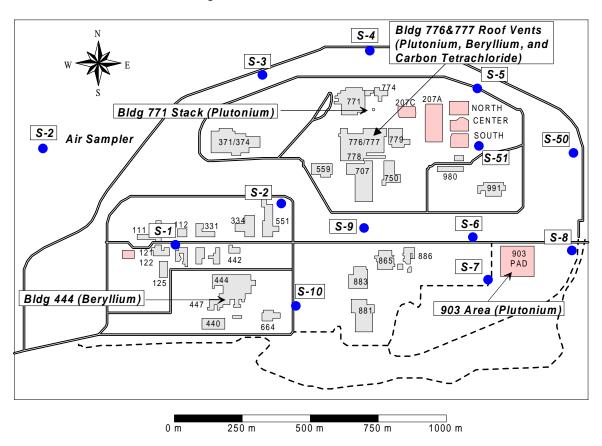
Some of the uncertainty in the beryllium and carbon tetrachloride slope factors was addressed by using a distribution of slope factors for the risk analyses. For example, for carbon tetrachloride we attempted to account for variation among cancer studies in rodents, metabolic conversions from animals to humans, body weight, and breathing rate variation. We considered uncertainty associated with assuming low dose linearity, extrapolating from high dose animal studies to low dose human exposures, variability in cancer response among animals and humans, and other sources of uncertainty, but these uncertainties could not be quantified.

The components of parameter uncertainty that appear in the cancer risk estimates express uncertainty in the source terms, environmental transport, and risk coefficients. Such components of uncertainty are real in the sense they can be derived from measured quantities or inferred from historical records. We did not include uncertainty related to the exposure scenarios in this assessment. The scenarios were constructed to represent hypothetical individuals living near the RFP during 1953–1989. Such scenarios make it possible to examine the sensitivity of risk estimates to different assumptions about location and time of exposure to RFP releases. The scenarios might also be helpful to some individuals whose exposure history appears to be similar

to the scenario. We emphasize that these scenarios only represent hypothetical individuals and do not correspond to any real person.

#### ATMOSPHERIC RELEASES AND RESULTING RISKS

This section describes the results of the in-depth evaluation of the estimated exposure and risks to the public from the key historical releases from the RFP. The Phase II findings confirmed that atmospheric releases of <u>plutonium</u> and <u>carbon tetrachloride</u> to the environment were the sources of the highest risks to those living offsite. Although of lesser significance, the risks associated with atmospheric <u>beryllium</u> releases are also presented because the same methodology was used for their evaluation as for plutonium and carbon tetrachloride. The potential significance of the surface water pathway to risk, which was reevaluated during Phase II, was minor compared to the impact of the air pathway. The atmospheric release points used in the model for these three contaminants are illustrated in Figure 3.



**Figure 3.** Atmospheric release points modeled for plutonium, beryllium, and carbon tetrachloride. Major fires occurred in Building 771 in September 1957 and in Buildings 776/777 in May 1969. The 903 Area was the outside storage location of drums of plutonium-contaminated waste oils. The large black circles indicate air sampler locations before 1973.

In our analysis, the risks from the different contaminants are reported separately; no attempt was made to combine the risk estimates. This approach was used because there is no established methodology for combining risks from radionuclides and chemicals. The current understanding of the interactions between contaminants in the human body and the resulting health risk is in its infancy. As a first step in determining the magnitude of the combined risk, the risk estimates might simply be added assuming they are additive. However, other risk levels are possible because of potential synergistic and antagonistic effects between different types of contaminants.

#### **Releases of Plutonium**

To understand the risks associated with atmospheric releases of plutonium from the RFP, it is convenient to divide the releases into a number of categories: routine operational releases, releases during the 1957 and 1969 fires, and releases from the 903 Area drum storage area. The history and details of the events are described here together with the release estimates. The risks associated with all the plutonium releases are discussed at the end of the section. A description of the modeling approach used for these releases is described in an earlier section of the report ("Modeling Approach").

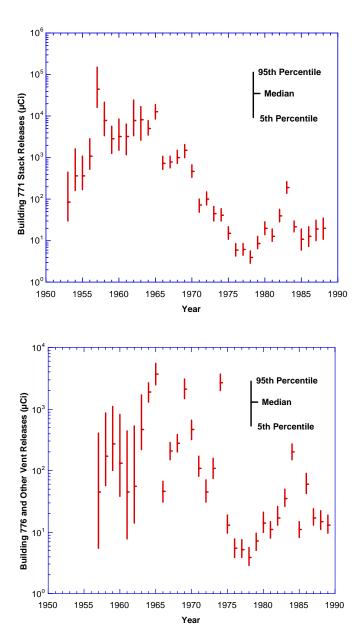
#### **Routine Operations**

Initially, Building 771, which became operational in 1953, housed essentially all of the plutonium operations. These included plutonium recovery and purification and plutonium component manufacturing (<a href="ChemRisk">ChemRisk</a> 1992). In 1958, many of the plutonium fabrication operations were moved from Building 771 to Buildings 776/777 when the building came online. The recovery operations stayed in Building 771 (<a href="ChemRisk">ChemRisk</a> 1992). Construction of Building 707, which was used for plutonium fabrication, was completed in 1972. Plutonium was released via the ventilation exhaust from these plant buildings as a result of normal routine operations at the RFP. Even though the ventilation exhaust systems included HEPA filters designed to remove the bulk of the plutonium, some plutonium was continuously released to the environment.

The quantity of plutonium routinely released from RFP buildings was estimated by ChemRisk® in Phase I of this study (ChemRisk 1994b). The amount released from routine operations was considerably less than from nonroutine events. The term routine releases is used to distinguish between releases from day-to-day operations and those from accidents that are considered separately. Included in the routine operational releases are many minor fires and other off-normal conditions that did not warrant separate analysis. During Phase II, the issues of nonrepresentative sampling of the plutonium concentration in the large exhaust ducts and of the self-absorption correction factor were evaluated by Voillequé (1999b). By combining these results with the bias and uncertainty estimates for the routine effluent monitoring data reported in Phase I, Voillequé (1999b) generated revised estimates of plutonium releases to the environment as a result of routine operations.

The median plutonium release estimate for the entire period of operations was approximately a factor of 3 larger than the Phase I estimate and had a broader range of uncertainty. Figure 4 shows the distributions of release estimates for Building 771 stack (upper graph) and for building roof vents (primarily Buildings 776/777 and 707) (lower graph). Before 1963, releases from the Building 771 stack dominated the routine plutonium emissions. The largest releases were

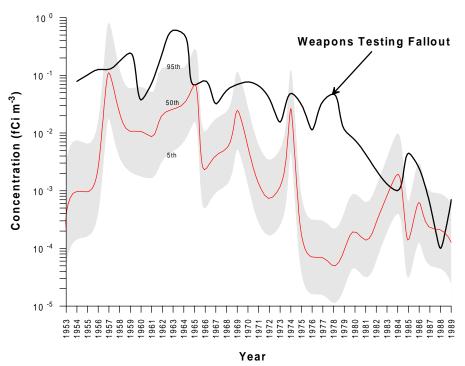
estimated to be in 1957, the year of the fire in Building 771. Release estimates for that year ranged from 14,000  $\mu$ Ci (5<sup>th</sup> percentile) to 130,000  $\mu$ Ci (95<sup>th</sup> percentile) and were due primarily to the extended cleanup work following the accident. The median annual release estimate for the Building 771 stack in 1989 was 0.06  $\mu$ Ci. Plutonium releases during the two major fires in 1957 and 1969 are not included in these estimates (they are described in the following sections). The uncertainty in the release quantity varied from year to year and was the greatest for the early years of plant operation.



**Figure 4.** Estimates of routine plutonium releases ( $\mu$ Ci). The upper graph shows the releases from Building 771 stack, and the lower graph the releases from Buildings 776/777 and other roof vents. Releases from the major fires in 1957 and 1969 are not included in the routine release estimates but are addressed separately below (source: Voillequé 1999b).

Effluent was passed through HEPA filtration before discharge to the atmosphere. The median particle size for HEPA filtered effluent was reported to be 0.3  $\mu$ m. However, when filter leakage occurred, larger particles that were more typical of the workplace aerosols would have been released. For this reason, routine operational releases were assumed to be characterized by a plutonium aerosol with an AMAD of ~1  $\mu$ m and a geometric standard deviation of 2.5 (Voillequé 1999b).

Figure 5 summarizes the estimated annual concentrations of plutonium in air east of RFP along Indiana Street (the location of highest concentration outside the buffer zone). The median estimates ranged from 0.1 fCi m<sup>-3</sup> in 1957 to  $5 \times 10^{-5}$  fCi m<sup>-3</sup> in 1978 (Rood 1999b). These values can be compared to weapons testing fallout concentrations of plutonium in air of 0.1 fCi m<sup>-3</sup> in 1957 and  $4 \times 10^{-2}$  fCi m<sup>-3</sup> in 1978. The geometric standard deviation of the estimated concentration of plutonium in air was typically around 2 to 2.4. A comprehensive analysis of the atmospheric dispersion and transport calculations for the routine releases is given in Rood (1999b) together with the exposure and risk calculations.



**Figure 5.** Estimated annual plutonium concentrations in air east of the RFP on Indiana Street outside the current buffer zone. The 50<sup>th</sup> percentile is shown as the center dark line in the shaded area that represents the 5th to 95th percentile range. Weapons testing fallout represents estimated <sup>239,240</sup>Pu concentrations in air from nuclear weapons tests as reported in Rope et al. (1999).

#### 1957 Fire

A major fire occurred in Building 771 on September 11, 1957, which resulted in a significant release of plutonium to the atmosphere. The fire began in a glove box at about 10 p.m., as a result of spontaneous ignition of metallic plutonium casting residues. Subsequently, the Plexiglas glove boxes caught fire and burned, as did the rubber gloves attached to the glove boxes. The firemen ultimately used water to extinguish the primary fire at 10:38 p.m.; however, the fire had already spread to the exhaust filter plenum system. At 10:39 p.m., there was an explosion in the ventilation system. Some plenum HEPA filters had burned through as early as 10:25 p.m., permitting the direct release of plutonium up the building stack. The exhaust filter plenum consisted of a long concrete-block walled room into which the individual exhaust systems discharged. Exhaust air was passed through a structural steel framework that contained 620, 61-cm (24-in.) square Chemical Warfare Service HEPA filters. Four exhaust fans were connected to the filtered side of the plenum and discharged into a common exhaust duct leading to a concrete tunnel and Building 771's 44-m (144-ft) high stack.

The largest releases of plutonium are estimated to have occurred between 10:15 p.m. and 10:45 p.m. because of the rapid burn-through of the booster system filters<sup>e</sup> and the main plenum filters before water was applied to the plenum filter fire. The exhaust fans were placed on high speed at 10:25 p.m. and stopped working at 10:40 p.m. when fire burned through the power cable. After 10:45 p.m., the main fire was out and releases were much lower. At 2:00 a.m., the fire was declared "knocked down," meaning that all obvious burning had been extinguished. By this time, water had been applied to the plenum fire for 2 hours and 45 minutes. The sum of all plutonium releases after 2:00 a.m. was a small fraction of the total estimated release.

A detailed analysis of the quantity of plutonium released during the fire is presented in <u>Voillequé</u> (1999c). Many factors and their uncertainties were taken into account. The different sources of plutonium that could have contributed to atmospheric releases during the fire were identified. These included the plutonium present in the fire area; plutonium that had accumulated on the booster system filters, main plenum filters, glove box exhaust filters, and room air prefilters before the fire; and plutonium deposited in ductwork that may have been suspended during the fire. Estimates of the collection efficiencies of the various filtration systems were made and supported by historic measurements of activity in plant exhaust systems.

A detailed analysis of the fire chronology established the probable sequence of events during the fire, described the progression of the fire through the filter systems, and provided input for the possible range of oxidation conditions that plutonium metal was exposed to during the fire (Diliberto 1999). The fire was modeled beginning at 10:00 p.m. on the evening of September 11, 1957, and ending at 2:00 a.m. the following morning of September 12, 1957. The first 45 minutes of the fire was modeled in 1-minute intervals to account for the rapidly changing conditions. Releases during this time were summed to give 15-minute totals. The remainder of the fire was modeled in 15-minute intervals, corresponding to the resolution of the available meteorological data.

<u>Table 3</u> summarizes the median estimate and the 5<sup>th</sup> and 95<sup>th</sup> percentiles of the distribution of release quantities for each 15-minute period during the event. The median estimate of the total

<sup>&</sup>lt;sup>e</sup> The name given to the glove box exhaust filters. This system employed eight Chemical Warfare Service filters in two stages to remove plutonium from the glove box exhausts and was located before the main filter plenum.

quantity of plutonium released was 290 g (21 Ci). The 5<sup>th</sup> and 95<sup>th</sup> percentiles of the distribution were 160 g (11 Ci) and 510 g (36 Ci), respectively. The release estimates were sensitive to the amount of vigorous oxidation of plutonium assumed, and there is considerable uncertainty that sufficiently high temperatures were achieved for this to occur. If vigorous oxidation did not occur in the manner assumed, the release estimate would be biased high.

Table 3. Estimated Plutonium Releases during the 1957 Fire (Voillequé 1999c)

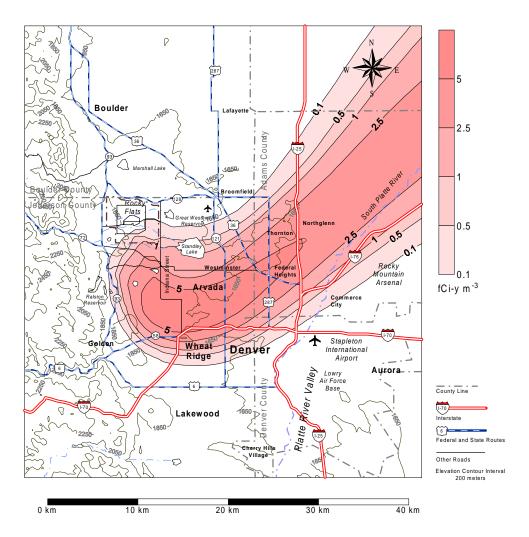
		Distribution of plutonium release quantities (Ci)		
Day	Time	5 <sup>th</sup> percentile	50 <sup>th</sup> percentile	95 <sup>th</sup> percentile
September 11	10:00–10:15 p.m.	0.034	0.06	0.099
	10:15-10:30 p.m.	5.7	11	17
	10:30-10:45 p.m.	4.1	8.8	16
	10:45–11:00 p.m.	0.14	0.28	0.50
	11:00-11:15 p.m.	0.14	0.27	0.49
	11:15–11:30 p.m.	0.067	0.14	0.26
	11:30–11:45 p.m.	0.067	0.13	0.25
	11:45-00:00 a.m.	0.067	0.13	0.25
September 12	00:00-00:15 a.m.	0.03	0.066	0.14
	00:15-00:30 a.m.	0.038	0.075	0.14
	00:30-00:45 a.m.	0.024	0.048	0.083
	00:45-01:00 a.m.	0.017	0.034	0.059
	01:00-01:15 a.m.	0.010	0.021	0.038
	01:15-01:30 a.m.	0.0069	0.015	0.027
	01:30-01:45 a.m.	0.0043	0.0095	0.019
	01:45-02:00 a.m.	0.0015	0.0044	0.011
Total		11	21	36

The size of plutonium particles released during the fire is not known. However, release estimates assumed all activity was respirable and could have ranged from submicron to up to  $10~\mu m$  AMAD. For this reason, we treated particle size stochastically in the transport calculations.

The plutonium released during the fire was modeled as puffs that entered the atmosphere every 15-minutes from 10:00 p.m. September 11 until 2:00 a.m. September 12, 1957. The transport calculations were continued until 7:00 a.m. to allow all the released plutonium to disperse throughout the model domain. The computer code simulations performed using RATCHET covered a 9-hour period. Because the effluent release temperature was estimated to be near 400°C (752°F), there was significant plume rise, and maximum plutonium concentrations in air 1 m (3.3 ft) above the ground were estimated some distance southeast of RFP, not adjacent to it. This represents the air concentration to which people would have been exposed.

At the time the fire started, the plume was transported in a westerly direction for a few kilometers. Around 10:45 p.m., the wind direction at the RFP shifted so that it blew out of the northwest and continued to blow from that direction until about 4:00 a.m., September 12. These winds transported the bulk of the airborne plutonium to Arvada and toward the Denver

metropolitan area. Near southern Arvada, the air mass converged with air flowing from the southwest in the Platte River Valley, which resulted in a northeasterly plume trajectory. Figure 6 shows the estimated plutonium concentrations in air (1 m [3.3 ft] above ground).



**Figure 6.** Estimated 9-hour average plutonium concentration in air 1 m (3.3 ft) above ground at the 50<sup>th</sup> percentile level during the 1957 fire.

#### 1969 Fire

A major fire occurred in a glove box in the north foundry glove box line in Buildings 776/777 on May 11, 1969. The fire started at about 2:00 p.m., when a pressed plutonium metal briquette, stored in an open can in the glove box, spontaneously ignited. At 2:27 p.m., the alarm was received at the fire station. Two minutes later, when the captain and three firemen arrived at the west-end of the building, there were flames 46 cm (18 in.) above the glove box line. At this time, the firemen reported two loud noises and observed fireballs, presumably because of the rapidly burning gases. Based on experience from the 1957 fire, the captain soon directed that water be used to fight the fire. The fire spread along the north foundry glove box line, but it was

prevented from moving into the north machining glove boxes by a barrier. The fire then spread along the north-south conveyer glove box line. The fire was observed in that area at 2:50 p.m., at which time a loud noise was heard and firemen felt vibrations on the second floor of the building. Considerable damage was done to the building and its equipment. Between 3:20 p.m. and 4:10 p.m., smoke was observed issuing from the roof and exhaust vents. The roof was sprayed with water and watched until after 5:00 p.m. The fire was considered contained by 6:40 p.m., and extinguished by 8:00 p.m., at which time a fire watch was established. Several small fires recurred during the night and the following morning.

During the early stages of the fire, Booster System No. 2, which serviced the north foundry line, became clogged. Air from this portion of the building was then processed through Booster System No. 1. Releases from the booster systems were monitored until 4:00 p.m., at which time the stack samplers were disabled by power failure. It was reported that the first two of the four HEPA filter banks in Booster System No. 1 were breached and that the gaskets in the third and fourth filter banks had also failed. The main exhaust system samplers, which operated between May 9 and May 15, indicated that approximately 2.8 mg (200  $\mu$ Ci) of plutonium was released via that exhaust. Measurements made following the fire of surface contamination on the roof indicated that most of the release was via the Booster System No. 1 exhaust duct. These measurements were central to estimating the magnitude of releases during the fire, which was modeled from 2:00 p.m. to 8:00 p.m., May 11, 1969. The total release, which was primarily from Booster System No. 1, is summarized in Table 4 and was estimated to be in the range 140 to 900 mg (0.01 to 0.06 Ci) of plutonium.

Table 4. Estimated Plutonium Releases during the 1969 Fire (Voillequé 1999d)

	Distribution of plutonium release quantities (mCi)		
Time	5 <sup>th</sup> percentile	50 <sup>th</sup> percentile	95 <sup>th</sup> percentile
2:00-3:00 p.m.	0.6	1.9	3.1
3:00–4:00 p.m.	2.6	7.5	12.4
4:00–5:00 p.m.	4.6	13.1	21.6
5:00–6:00 p.m.	3.3	9.3	15.5
6:00–7:00 p.m.	1.3	3.7	6.2
7:00–8:00 p.m.	0.6	1.9	3.1
Total	13	37	62

There were no measurements of the particle size of the plutonium released during the fire. However, the HEPA filters in the main filter plenum and booster systems were largely intact. Therefore, it was assumed that most of the particles released were relatively small and respirable. Significant deposition of larger particles on the roof and nearby ground surfaces was likely because the main discharge was via a duct that faced down toward the roof of the building.

We modeled the plutonium released during the fire as puffs that entered the atmosphere every 15-minutes after 2:00 p.m. until 8:00 p.m. May 11, 1969. The transport calculations were continued until 5:00 a.m. the following morning to allow all the released plutonium to disperse throughout the model domain. The RATCHET simulations covered a 15-hour period.

The plume of contamination was initially transported west of the RFP by the prevailing easterly winds that occur during upslope conditions. This continued until about 10:00 p.m., when

there was a change to downslope conditions and a reversal of the plume trajectory to the east. The largest releases are estimated to have occurred from about 3:00 p.m. to 6:00 p.m. The highest 15-hour plutonium concentrations in air (0.67 pCi m<sup>-3</sup> at the 95<sup>th</sup> percentile) were estimated at the west entrance of the RFP.

#### 903 Area

The 903 Area is located in the eastern part of the main production area of the RFP (Figure 3), and during the late 1950s and 1960s it served as a waste storage area for barrels containing plutonium-contaminated cutting oil and degreasing agents. Carbon tetrachloride was the primary degreasing agent. The combination of radioactivity and hazardous chemicals precluded its storage in traditional waste disposal locations at the RFP. Consequently, the waste was transferred to 55-gallon barrels that were stored outside on a grassy area that became known as the 903 Area. More than 4500 barrels were reported to have been stored at the 903 Area.

Site personnel reported barrel corrosion and subsequent leakage onto the soil as early as 1962. By 1964, this had become a large-scale issue, and fences were constructed to limit the spread of contamination by intruding wildlife. In 1967, efforts were made to remove the barrels from the 903 Area, repackage the contents, and ship the waste offsite to the National Reactor Testing Station, an Atomic Energy Commission facility near Arco, Idaho. Barrel removal was completed in June of 1968. However, by late 1968, there was evidence that wind action had transported plutonium-contaminated soil beyond the 903 Area. In 1969, the area was paved with asphalt to prevent further transport of plutonium-contaminated soil away from the area. The first coat of asphalt was placed over the contaminated area in July 1969, although paving was not completed until November of that year. However, high wind events during January had already spread the contamination further, as did the surface grading and leveling operations that took place in March and April in preparation for paving. The mass of plutonium released to the soil from the leaking barrels is estimated to range between 85 g (6.1 Ci) and 800 g (58 Ci) (Meyer et al. 1996).

We calculated atmospheric releases from the 903 Area for a 6-year period, 1964 through 1969, and divided them into two types: discrete event releases and continuous releases. Discrete event releases occurred during short-term high-wind events in 1968 and 1969 and were modeled as individual days. In contrast, continuous releases occurred during more typical meteorological conditions and were calculated annually from 1964 to 1969.

Air monitoring data from sampler S-8 (Figure 3) were used to identify discrete event releases. This was possible because the S-8 sampler was located east of the 903 Area in the path of soil particles suspended by wind storms. The wind usually blows from west to east during such storms. A total of 24 discrete release event days were identified. Meteorological data for these 24 days were obtained from a portable meteorological station, designated Kent, which was located about 10 km (~6 mi) north of RFP. The station was operated by the National Center for Atmospheric Research in Boulder. As the releases were further investigated, we determined that 6 of the 24 days accounted for about 90% of the total releases (Weber et al. 1999). These 6 days were modeled as discrete events, and the remaining 18 days were included with the continuous plutonium releases.

<sup>&</sup>lt;sup>f</sup> Current name, Idaho National Engineering and Environmental Laboratory (INEEL).

The uncertainty estimates for the amount of plutonium released into the atmosphere from the 903 Area during the 6-year period for discrete and continuous releases are summarized in Tables  $\underline{5}$  and  $\underline{6}$ . The median estimate for the total activity of plutonium released on particles <30  $\mu$ m aerodynamic equivalent diameter (AED) was 3.1 Ci (43 g), and the 5<sup>th</sup> and 95<sup>th</sup> percentiles of the distribution were 1.4 Ci (19 g) and 15 Ci (208 g), respectively. Releases of plutonium attached to particles >30  $\mu$ m AED were not quantified because the S-8 sampler was not capable of detecting these large particles. While these large particles did not pose a direct inhalation health risk, they did contribute to offsite contamination.

The six discrete release events resulted in relatively narrow plumes of airborne contamination that extended in an easterly direction from the 903 Area but with a more pronounced northeast-southwest elongation. These high wind events were responsible for suspension of relatively large amounts of plutonium-contaminated soil. For example, east of the plant along Indiana Street, they accounted for about 50% of the airborne plutonium activity concentration (~1 fCi-y m<sup>-3</sup>). However, by their very nature, high wind events also resulted in greater dilution, dispersion, and depletion within the airborne plume than would be estimated had the same activity been released over a longer period of time and modeled using annual average meteorological data.

Table 5. 903 Area Discrete Release Estimates for Plutonium Attached to <30 µm AED Soil Particles (Weber et al. 1999)

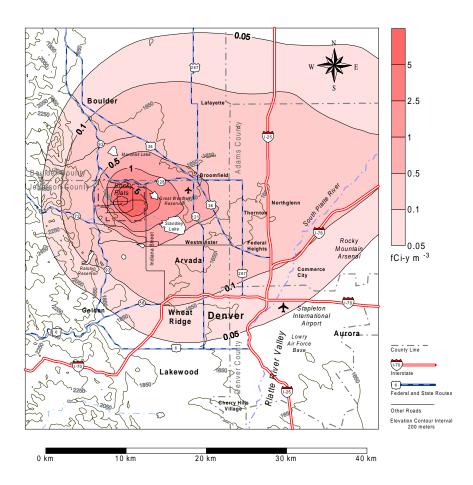
•		·		
	Distribution o	Distribution of estimated release quantity (Ci)		
Release date	5 <sup>th</sup> percentile	50 <sup>th</sup> percentile	95 <sup>th</sup> percentile	
5-Dec-68	0.0097	0.051	0.25	
6-Jan-69	0.034	0.18	0.86	
7-Jan-69	0.18	0.89	4.3	
30-Jan-69	0.096	0.53	2.4	
19-Mar-69	0.012	0.055	0.27	
7-Apr-69	0.077	0.38	1.9	
Total <sup>a</sup>	1.2	2.8	14	

<sup>&</sup>lt;sup>a</sup> The distribution of total releases was determined by sampling from the distribution of each individual release event and summing. It is not the sum of the given percentile value.

Table 6. 903 Area Continuous Release Estimates (1964–1969) for Plutonium Attached to  $<30~\mu m$  AED Soil Particles (Weber et al. 1999)

	Distribution of	Distribution of estimated release quantity (Ci)			
Year	5 <sup>th</sup> percentile	50 <sup>th</sup> percentile	95 <sup>th</sup> percentile		
1964	0.00081	0.0066	0.038		
1965	0.00094	0.013	0.10		
1966	0.0013	0.014	0.11		
1967	0.0030	0.026	0.16		
1968	0.039	0.19	1.0		
1969	0.022	0.11	0.55		

In contrast, the continuous releases resulted in an east-southeast trending ellipsoid shaped plume that reflects the predominant wind direction from the west-northwest. Higher concentration isopleths near the source trend mostly easterly; however, farther away from the source, they trend to the northeast. The northeast trend is believed to be due to the influence of the Platte River Valley and the diurnal pattern of upslope-downslope conditions that characterize the general air movement on the Colorado front range environs. The estimated time-integrated concentration of plutonium attached to respirable soil particles ( $<15~\mu m$  AED) for all 903 Area releases (discrete and continuous) is shown in Figure 7.



**Figure 7.** Ground-level time-integrated concentration of plutonium at the  $50^{th}$  percentile level for respirable particles (<15  $\mu$ m AED) released from the 903 Area between 1964 to 1969.

The percentage of particles comprising three different particle size fractions was considered for the atmospheric transport modeling:  $<3~\mu m$  AED,  $3-10~\mu m$  AED, and  $10-15~\mu m$  AED. The organ-specific plutonium risk coefficients for plutonium aerosols with AMADs of  $1~\mu m$ ,  $5~\mu m$ , and  $10~\mu m$  were applied to the three size fractions, respectively. Particles  $>15~\mu m$  AED were not considered in the atmospheric transport modeling because they are not considered respirable and do not penetrate as far as the bronchial region of the respiratory tract. They are eliminated from the body either by direct expulsion (e.g., nose blowing) or via transfer to the gastrointestinal tract.

# **Summary of Plutonium Releases**

Distributions of the total estimated plutonium release quantities for the four categories of release events evaluated in Phase II are shown in Table 7. The 1957 fire and 903 Area releases dominate the total estimated quantity of plutonium released from the site. The duration of the release events varied from a period of hours for the two fires; to about 6 years for the 903 Area (with the major releases occurring on 6 days); to a period of several decades for the routine operational releases. The plutonium particle size distributions also varied for the different release events. Releases from routine operations and the 1969 fire were characterized by small particle sizes (<3  $\mu m$  AED). Releases during the 1957 fire were also assumed to contain predominantly small particles with the possibility that larger particles up to 10  $\mu m$  AED were released. Releases from the 903 Area were characterized by a wide range of particle sizes, and particles up to 30  $\mu m$  AED were accounted for in the analysis. However, only those <15  $\mu m$  AED were assumed to be respirable. At ground level, the plume of plutonium particles released from the 1957 fire was confined to a relatively small area in the model domain (Figure 6). Furthermore, ground deposition of plutonium particles from the 1957 fire was less than for the 903 Area releases because the particle sizes tended to be much smaller, yielding lower settling velocities.

Distribution of estimated release quantity (Ci) 5<sup>th</sup> percentile 50<sup>th</sup>percentile 95<sup>th</sup> percentile Release event Routine operations 0.087 0.12 0.25 1957 fire 11 21 36 1969 fire 0.013 0.037 0.062

3.1

15

**Table 7. Plutonium Release Estimate Distributions by Event** 

1.4

### **Risks from Plutonium Releases**

903 Area<sup>a</sup>

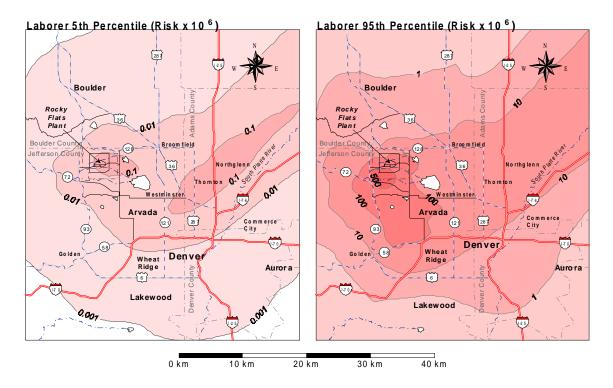
The lifetime cancer incidence risks from exposure to all plutonium releases that occurred from 1953 through 1989 are presented for the laborer scenario because this was the hypothetical scenario that resulted in the largest risk estimates. The laborer had the highest risk of all scenarios because he was assumed to have lived in the model domain for the entire period the RFP operated (1953–1989) and had the highest breathing rate of any of the hypothetical individuals.

The spatial distribution of the lifetime cancer incidence risk for the four main cancer sites (lung, liver, bone surface, and bone marrow) for plutonium inhalation for the laborer scenario is shown in Figure 8. These four sites account for ~97% of the total risk from plutonium inhalation (Grogan et al. 1999). The individual organ risk was highest for the lung. The median lung cancer risk was about a factor of 2 larger than the median liver cancer risk, which was about a factor of five larger than the bone cancer risk. The median leukemia risk was the smallest and was almost two orders of magnitude lower than that for lung cancer (see also Tables 1 and 2).

The two isopleth maps in <u>Figure 8</u> show the 5<sup>th</sup> and 95<sup>th</sup> percentile values of the output distributions. These percentiles represent the 95% confidence interval around the 5<sup>th</sup> and 95<sup>th</sup> percentile values (Rood and Grogan 1999d). The area of maximum risk at the 95<sup>th</sup> percentile level

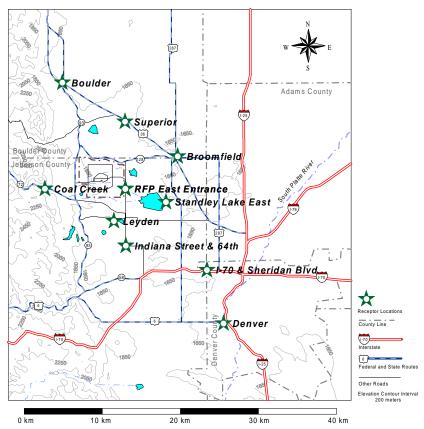
 $<sup>^{</sup>a}$  Includes particles up to 30  $\mu$ m AED; ~20% were estimated to be in the respirable size fraction (<15  $\mu$ m AED).

is represented by the 100 contour in Figure 8, and extended south of the RFP to the intersection of Colorado 58 and Interstate 70. These maximum incremental lifetime cancer incidence risks were in the  $10^{-4}$  range (1 chance in 10,000) of developing cancer during a lifetime. However, a single grid node near the southwest corner of the RFP boundary had a 95<sup>th</sup> percentile cancer risk value of  $1.1 \times 10^{-3}$  (about 1 chance in a 1000). The spatial extent of this excursion above the EPA's acceptable risk range was limited to an area no greater than 1 km² that historically was uninhabited. At the 5<sup>th</sup> percentile level, the maximum cancer risk was in the  $10^{-7}$  range (1 chance in 10 million) of developing cancer during a lifetime.



**Figure 8.** Lifetime cancer incidence risk from plutonium inhalation for the laborer scenario;  $5^{th}$  percentile (left plot) and  $95^{th}$  percentile (right plot). Risk values have been multiplied by  $10^6$  so a value of 1.0 in the plot represents a cancer risk of  $1 \times 10^{-6}$  or 1 chance in 1 million of developing cancer.

To demonstrate the importance of location and time of exposure, we extracted cancer risk estimates by decade of exposure since plant operations began in 1953 at selected locations in the model domain (Figure 9) for the laborer scenario (Figure 10). The relative importance of each decade of exposure depended on the location within the model domain and the percentile level chosen. The maximum risks at the 95<sup>th</sup> percentile were estimated from exposure during the years 1953–1959, provided the laborer was located in the path of the plume from the 1957 fire. The selected locations outside the plume from the 1957 fire were Denver, Boulder, Broomfield, Superior, and RFP East Entrance.

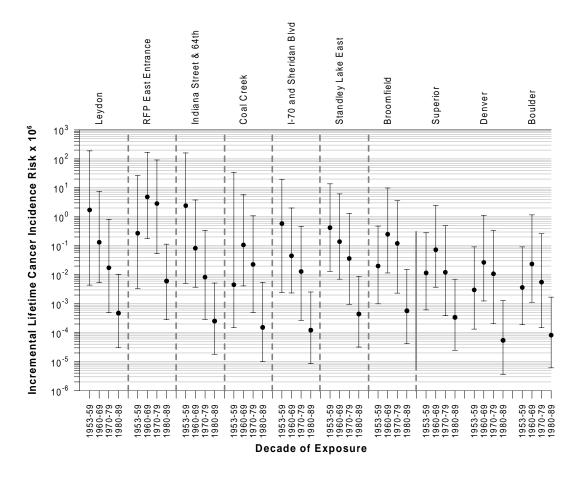


**Figure 9.** Map showing assumed locations for the laborer scenario used to evaluate cancer risk.

Cancer risk estimates at these and other locations outside the 1957 plume path were dominated by exposure during the 1960s, primarily from 903 Area releases. In most cases, the 50<sup>th</sup> percentile and 5<sup>th</sup> percentile level followed this trend. One exception was the Coal Creek location (Figure 10), which lies to the east of the RFP. The maximum risk at the 50<sup>th</sup> percentile level was estimated from exposures in the 1960s; at the 95<sup>th</sup> percentile level, the maximum risk was estimated from exposures in the 1950s. There is a large uncertainty in the estimated risk from exposure during the 1950s for the laborer located at Coal Creek. This uncertainty reflects the uncertainty in the 1957 fire plume path. While there is a low probability the plume traveled in that direction, the resulting risks are relatively high.

Lifetime cancer risk estimates for the laborer scenario at the receptor locations discussed above are tabulated for the 5<sup>th</sup>, 50<sup>th</sup>, and 95<sup>th</sup> percentiles in <u>Table 8</u>. Using the laborer located at Leyden as an example, the cancer risk estimates can be interpreted as follows:

- There is a 90% probability that the model estimated incremental lifetime cancer incidence risk for the laborer located in Leyden was between  $2.2 \times 10^{-8}$  (5% value) and  $2.0 \times 10^{-4}$  (95% value)
- There is a 5% probability that the model estimated incremental lifetime cancer incidence risk for the laborer located in Leyden was greater than  $2.0 \times 10^{-4}$
- There is also a 5% probability the model estimated risk was less than  $2.2 \times 10^{-8}$ .



**Figure 10.** Lifetime cancer incidence risk from plutonium inhalation for the laborer scenario at selected locations in the model domain. Dots represent the  $50^{th}$  percentile value; horizontal bars represent the  $5^{th}$  and  $95^{th}$  percentiles. Cancer risks have been sorted by decade of exposure.

We may also interpret this to mean, given an exposure history and lifestyle similar to that of the laborer scenario residing in Leyden, there is a 50% probability that there would be about 2 cases of cancer per million similarly exposed individuals, and because of the large uncertainty, a 5% probability that the estimated number of cancer cases attributed to inhalation of plutonium originating from the RFP would be greater than 200 cases per million people at that location, or a 5% probability that there may be as few as 0.02 cases in a population of 1 million similarly exposed individuals. Estimated cancer risks at the 95<sup>th</sup> percentile level are within the point of departure for acceptable risks established by the EPA of 10<sup>-6</sup> to 10<sup>-4</sup>.

Denver

Boulder

Coal Creek

Indiana Street and 64th

Incremental lifetime cancer incidence risk <sup>a</sup> 50<sup>th</sup> percentile 95<sup>th</sup> percentile 5<sup>th</sup> percentile Receptor location **RFP East Entrance** 0.53 11 270 3.2 **Superior** 0.0069 0.11 Broomfield 0.021 0.46 20 Stanley Lake East 0.045 0.77 27 I-70 and Sheridan Blvd 0.019 0.72 24 2.3 200 Leyden 0.022

0.05

0.042

0.2

2.6

1.6

1.3

59

150

Table 8. Lifetime Cancer Incidence Risk from Plutonium Inhalation for the Laborer Scenario at Selected Receptor Locations in the Model Domain

0.0026

0.0023

0.01

0.026

### **Releases of Carbon Tetrachloride**

The source term and estimated uncertainty for carbon tetrachloride compiled by ChemRisk® during Phase I was investigated in Phase II and documented in McGavran and Rood (1999b). We did not find any additional information regarding the amounts used or significant process changes that could help refine the release estimates. Investigation of amounts shipped in waste to Idaho and in the aquifer underlying the RFP confirmed the Phase I conclusion that the vast majority of carbon tetrachloride used at the RFP was released to the atmosphere.

Carbon tetrachloride is a volatile organic compound and calculations showed almost all the compound that was disposed of on the ground surface (a common disposal practice in the past) would be released to the air. We assumed most of the carbon tetrachloride routinely used at the RFP evaporated during cleaning and degreasing operations and was vented to the atmosphere via roof vents on Buildings 776/777 and 707 where operations of this type were performed. Release estimates were made for three time periods and are summarized in Table 9. The estimates were based on inventory amounts, plutonium production, and solvent degreaser use during the different time periods (ChemRisk 1994b). In total, 1100 to 5400 tons of carbon tetrachloride was estimated to have been released from 1953–1989 (McGavran and Rood 1999b). This release range was described by a uniform distribution (Table 9).

**Table 9. Carbon Tetrachloride Source Term Estimates** 

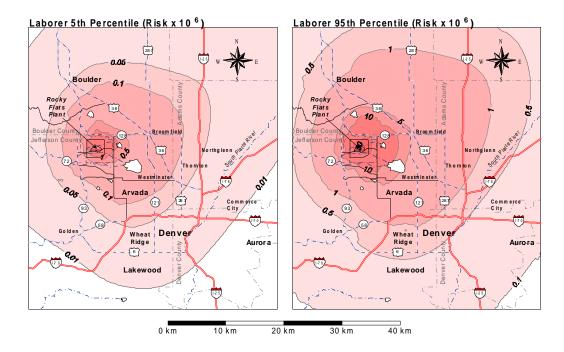
Time period	Estimated release in tons per year <sup>a</sup>
1953–1957	4–20
1958-1970	40–200
1971–1989	40–200 in 1970, decreasing linearly to 20–100 by 1989.
<sup>a</sup> Lower and upper bound	d estimates

<sup>&</sup>lt;sup>a.</sup> Risk values have been multiplied by  $1 \times 10^6$  thus the tabled value of 11 represents a cancer risk of  $11 \times 10^{-6}$  or 11 chances in 1 million of developing cancer.

The maximum estimated air concentrations of carbon tetrachloride along Indiana Street for 1988 was 0.19  $\mu g$  m<sup>-3</sup>. Concentrations for 1988 ranged from 0.095 (5<sup>th</sup> percentile) to 3.2  $\mu g$  m<sup>-3</sup> (95<sup>th</sup> percentile). These air concentrations can be compared to typical background concentrations in rural areas of 1  $\mu g$  m<sup>-3</sup>. Geometric mean incremental lifetime cancer incidence risk estimates for carbon tetrachloride inhalation (Figure 11) were greatest for the laborer scenario (5.2 × 10<sup>-6</sup>, GSD = 2.4).

The risks may be interpreted as follows:

- There is a 90% probability that the model-estimated incremental lifetime cancer incidence risk for the laborer was between  $1.3 \times 10^{-6}$  (5% value) and  $2.1 \times 10^{-5}$  (95% value)
- There is a 5% probability that the model-estimated incremental lifetime cancer incidence risk for the laborer was greater than  $2.1 \times 10^{-5}$
- There is also a 5% probability the model-estimated incremental lifetime cancer incidence risk for the laborer was less than  $1.3 \times 10^{-6}$ .



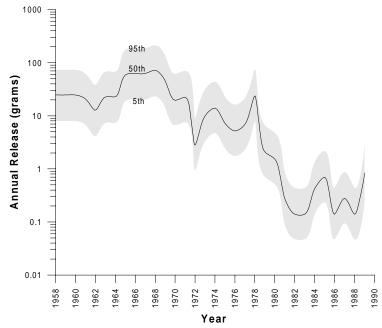
**Figure 11.** Lifetime cancer incidence risk from carbon tetrachloride inhalation for the laborer scenario;  $5^{th}$  percentile (left plot) and  $95^{th}$  percentile (right plot). Risk values have been multiplied by  $10^6$  so a value of 1.0 in the plot represents a cancer risk of  $1 \times 10^{-6}$  or 1 chance in 1 million of developing cancer. The risk is based on cumulative inhalation of carbon tetrachloride for the years 1953-1989.

### **Releases of Beryllium**

Although beryllium was initially used in research and development at the RFP in 1953, beryllium operations only became significant with respect to potential releases beginning in 1958. Beryllium manufacturing operations in Building 444 included casting (foundry), cutting, heat-

treating, rolling, and machining. Beryllium foundry operations ceased in 1975. Details of beryllium component manufacturing, machining, cutting, heat treating, and rolling and other operations and ventilation systems used to control beryllium emissions over the years are described in Phase I and II technical reports (<u>ChemRisk</u> 1992; <u>McGavran and Rood</u> 1999a). With the possible exception of effluent from one building in the early 1960s, all air exhaust discharged from RFP beryllium-processing facilities was subjected to HEPA filtration that was installed to control particulate radioactive effluents (<u>ChemRisk</u> 1992).

Beryllium has been monitored in the plant air exhaust effluent since at least 1963. Air exhaust samples were taken from filter plenum exhausts after the air passed through HEPA filters but before it exited the stack. The sampling practices, sampling system design, sample line losses, calculations of flow rates, and exhaust volume and uncertainties determined previously for radioactive particles were applied to the beryllium sampling data (ChemRisk 1994b). Figure 12 summarizes the estimates with uncertainties for beryllium releases to air. Median release estimates were typically around  $10-30 \text{ g y}^{-1}$  for the years 1958 to 1971 and generally less than  $10 \text{ g y}^{-1}$  after 1971. The median total release from 1958 to 1989 was 324 g. The largest release occurred in the year 1968. Beryllium was also released during three fires that occurred in 1962, 1964, and 1978. These releases were monitored by the stack sampling equipment and, therefore, were included in the annual release estimates.



**Figure 12.** Annual release estimates for beryllium with uncertainty as estimated by <u>ChemRisk</u><sup>®</sup> (1994b). The 50<sup>th</sup> percentile values are shown as the dark line in the shaded region representing the 5<sup>th</sup> to 95<sup>th</sup> percentile range.

The median estimated concentrations of beryllium in air at the location of highest concentration outside the buffer zone (east of the plant along Indiana Street) for all years in the assessment ranged from  $1.3 \times 10^{-6}$  ng m<sup>-3</sup> in 1986 to  $7.3 \times 10^{-4}$  ng m<sup>-3</sup> in 1968, the year of highest release. The maximum concentration in the model domain for 1968 was calculated within

the plant buffer zone and ranged from  $2.5 \times 10^{-3}$  ng m<sup>-3</sup> (5<sup>th</sup> percentile) to  $6.8 \times 10^{-2}$  ng m<sup>-3</sup> (95<sup>th</sup> percentile). This can be compared with an annual average natural background range of 0.03 to 0.3 ng m<sup>-3</sup> (median of  $1 \times 10^{-1}$  ng m<sup>-3</sup>) estimated in Rope et al. (1999).

Incremental lifetime cancer incidence risk estimates for beryllium inhalation were greatest for the laborer scenario  $(3.9 \times 10^{-10})$ . These risks may be interpreted as follows:

- There is a 90% probability that the model-estimated incremental lifetime cancer incidence risk to the laborer was between  $7.5 \times 10^{-11}$  (5% value) and  $1.8 \times 10^{-9}$  (95% value)
- There is a 5% probability that the model-estimated incremental lifetime cancer incidence risk for the laborer was greater than  $1.8 \times 10^{-9}$
- There is also a 5% probability the model-estimated risk was less than  $7.5 \times 10^{-11}$ .

Although beryllium exposures for workers at the RFP have been of great concern and the attention to workers may have caused public concern about health effects because of beryllium exposure offsite, the results of this assessment estimated that lung cancer risk from beryllium exposures offsite were negligible.

Great uncertainty is associated with estimating the risk for chronic beryllium disease in the offsite public. The maximum estimated beryllium concentration in air in the entire model domain ranged from  $2.5 \times 10^{-6}$  to  $6.8 \times 10^{-5} \, \mu g \, m^{-3}$  onsite, concentrations more than 300 times less than the EPA's reference concentration (RfC) of  $2.0 \times 10^{-2} \, \mu g \, m^{-3}$ . The maximum air concentration estimated along Indiana Street ranged from  $9.4 \times 10^{-7}$  to  $1.4 \times 10^{-5} \, \mu g \, m^{-3}$ , concentrations more than 1400 times less than the RfC. A hazard index calculated using these values would be well below 1; therefore, any cases of chronic beryllium disease are unlikely. However, because of the complexity and apparent immunological nature of chronic beryllium disease, it is difficult to conclude that no cases of chronic beryllium disease may have occurred from offsite exposure.

### ESTABLISHING CONFIDENCE IN THE RESULTS

In this study we used models extensively to estimate the risks to people from plutonium, carbon tetrachloride and beryllium released to the atmosphere from the RFP. The choice of models was based on the available data, the required resolution of the calculations, and uncertainty considerations. Throughout this project, every effort has been made to validate the models to establish confidence in the estimated magnitudes and impacts of the contaminant releases. An important part of the model validation process involves comparing model estimates against an independent set of measurements of like quantities. Model comparison is the means by which the accuracy of the model is evaluated. Accuracy is a measure of how close the model estimation is to a similar measured quantity. We contrast this with model precision, which is the ability of the model to reproduce the same result upon repeated trials, given uncertainty in the input parameters. The parameter uncertainty analysis discussed earlier was used to establish model precision. Other aspects of model validation include testing the face validity of the models and testing the input assumptions. Face validity addresses the acceptability of the model structure and parameters to others familiar with the system. Peer review of the models and assumptions by established experts in the fields of application was used to address these aspects. The HAP and public also participated in the review process at all stages of the project.

Environmental monitoring data potentially useful for model validation were compiled and evaluated in Rope et al. (1999). The validation effort focused on plutonium because quality

environmental measurement data were lacking for carbon tetrachloride and beryllium. The same models were used to estimate the atmospheric release and transport of carbon tetrachloride and beryllium as for routine releases of plutonium; therefore, the air monitoring validation data compiled for plutonium are relevant for these other materials released. This summary of the validation for plutonium transport in the environment gives us some confidence that we have estimated the transport of other contaminants reasonably well. Environmental measurements include plutonium concentrations and total long-lived alpha activity in soil, vegetation, ambient air, water, and lake sediment. Other measurements include plutonium deposition flux and miscellaneous plutonium measurements in lichens, human tissue, human urine, and cattle. The model used in our analysis provided estimates of plutonium concentrations in ambient air and soil. Additional modeling and assumptions were necessary to estimate concentrations in other environmental media. In general, we limited our comparisons to measurements in soil, ambient air, lake sediment, and vegetation.

It is important that the environmental data sets used in model validation exercises are independent of data sets used to develop and calibrate the model. Therefore, it is not valid to compare estimated concentrations with measured values that were originally used to calibrate the model. For example, the source term for the 903 Area was developed and calibrated using measurement data from the S-8 air sampler (Figure 3). Therefore, comparisons of model estimated air concentrations with S-8 air sampler data do not provide evidence of model validation. It is also important that the measured quantities selected for comparison are within the same time and spatial resolution as the model. Spatial resolution of the model was 1 km (0.6 mi) and temporal resolution was typically 1 year. However, provisions were made in the code to provide concentrations averaged over the duration of a discrete event that ranged from 9 to 26 hours.

We acknowledge that we used portions of two soil data sets (Webb 1996; Poet and Martell 1972) to develop plutonium depth distributions. Consequently, these data sets are not entirely independent. Nevertheless, uncertainties associated with the source term, transport, and deposition of plutonium in airborne plumes far outweigh the uncertainties associated with the depth distribution. The model comparisons of plutonium soil inventories are completely independent (because depth distribution is not considered) and provide perhaps the best validation of total plutonium quantities released from the RFP. All the model comparisons were qualitative. That is, we did not perform any statistical analysis between the estimated and observed values or base the performance of the model on some numeric criteria.

## **Comparisons with Soil Measurements**

Soil represents a significant sink for plutonium deposited from the air. Plutonium is generally highly insoluble and becomes attached to soil particles. For those reasons, it accumulates in the upper soil layers. Measurements of plutonium concentrations in soil were useful for validating the total plutonium released from the site and, in particular, releases from the 903 Area because the 903 Area is believed to have been responsible for most of the offsite plutonium contamination from the RFP releases. (See the discussion at the end of this section.)

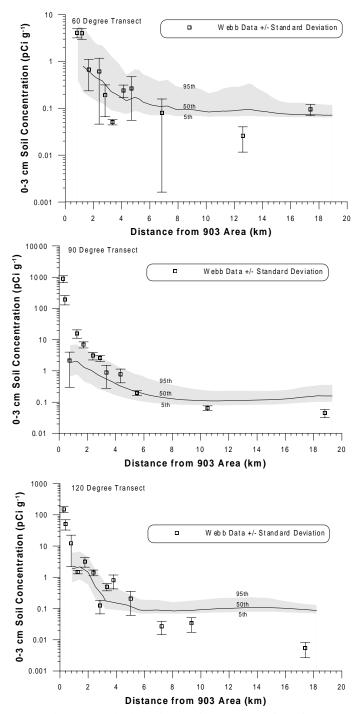
Few measurements of plutonium concentrations in soil were performed before 1969. Any information we found referred to total alpha activity in soils from scattered locations. Several technical problems were identified with comparing soil measurements to model estimates. First,

studies used different sampling depth and sample collection methods. The sampling depth used in soil studies varied from several millimeters to ~20 cm (~8 in.). The sampling depth and fraction of the total deposited activity in that depth must be known for a valid comparison to be made between estimated and measured soil concentrations. Second, most of the studies we reviewed did not systematically measure plutonium in soil with distance and direction from the 903 Area. Systematic sampling is important for evaluating the spatial trend of plutonium concentrations in soil.

The Webb (1996) data set was identified as the best available source of soil model validation data. Webb (1996) measured plutonium concentrations in soil in 1994, along four transects out to distances of ~20 km (~12 mi). A consistent protocol was used for all samples, and consideration was given to soil density, rock volume, and distribution with depth. Webb (1996) also provided inventory estimates with uncertainty within his study domain, which covered 209 km² (81 mi²).

We extracted estimated plutonium concentrations in the 0–3-cm (0–1.2-in.) soil layer for 1989 from the model output by interpolating concentrations between each of the output nodes along each of Webb's transect lines. Model-estimated soil concentrations for 1994 were not available because the model was not set up to make computations beyond 1989. However, the estimated concentrations in 1994 would essentially be the same. The nearest node was located about 500 m (1640 ft) east of the 903 Area. Consequently, estimated concentrations within 500 m (1640 ft) of the 903 Area were unavailable. Estimated plutonium concentrations in soil were extracted separately for the 5<sup>th</sup>, 50<sup>th</sup>, and 95<sup>th</sup> percentile values and are shown in Figure 13 along with the corresponding measured values. We added a mean fallout plutonium concentration of 0.06 pCi g<sup>-1</sup> determined by Webb (1996) to all the model estimated values because the measurements include plutonium from RFP and fallout sources.

Estimated soil concentrations 2 to 10 km (1.2 to 6 mi.) east of the 903 Area were generally within the range of measured values. At distances <2 km (1.2 mi) from the 903 Area, the model tended to underestimate concentrations, while at distances >10 km (6 mi) the model overestimated concentrations. Model bias was most clearly seen in the 90° and 120° sampling transects used by Webb (1996). Much of the model bias was attributed to the particle size distribution of 903 Area releases. Modeled releases from the 903 Area did not include contaminated soil particles >30 µm AED. These particles are not an inhalation health risk because they are not respirable; however, they do contribute to offsite contamination. Overall, the distribution assigned to particles <30 µm AED appeared to have been biased toward the finer particles. That is, a greater fraction of the activity was assumed to be associated with smaller particles than the soil concentration measurements suggest. Larger particles would deposit closer to the source, resulting in higher estimated soil concentrations close to the source (the 903 Area) and lower estimated soil concentrations at greater distances. If the particle size distribution had been calibrated using the observed deposition pattern, the estimates of plutonium inhalation from 903 Area releases would have been lower because a greater fraction of the activity would be associated with larger, nonrespirable particles.



**Figure 13.** Estimated and measured plutonium soil concentrations in the 0–3-cm (0–1.2-in.) layer for three transects originating from the 903 Area and extending eastward. Estimated results include 0.06 pCi g<sup>-1</sup> from fallout. Measured data are from Webb (1996) and represent plutonium from RFP sources and fallout. The 50<sup>th</sup> percentile values are shown as the dark line in the shaded region that represents the 5<sup>th</sup> to 95<sup>th</sup> percentile range.

Soil inventory refers to the total amount of plutonium in the soil column in a defined area. We compared estimated soil plutonium inventories with inventory estimates made by Webb and other researchers and reported in <a href="Webb">Webb</a> (1996). The inventory estimates were based on extrapolation of measured plutonium concentrations in soil. Inventories were reported for the Webb study area and for the total area affected by the RFP plutonium. The Webb study area was bounded by the 60° and 120° transects out to a distance of about 20 km (12 mi) from the 903 Area and covered 209 km²(81 m²). Model-estimated inventories for the Webb study area were generated by integrating estimated surface concentrations over an area bounded by the 60° and 120° transects. The total area affected by the RFP plutonium varied among researchers, but it was estimated to range from 600 to >2000 km²(230 to >770 mi²). Our calculations assumed the entire model domain (2200 km² [850 mi²]) was the total affected area. However, most of the plutonium in soil was restricted to an area of about 1000 km² (390 mi²) where deposition from the 903 Area releases occurred.

The inventory estimates represented only the RFP plutonium sources. Webb's inventory estimates within his study area were generally lower than those made by other researchers. This was attributed to the exclusion of rock volume in estimating inventories and other factors including depth distributions, soil density, and contributions from fallout sources of plutonium. Comparisons of inventory estimates are shown in Figure 14. Our estimate of the plutonium inventory for the Webb study area (upper graph in Figure 14) was slightly less than that made by Webb, but the two are in general agreement. Some of the difference was attributed to underestimating plutonium soil concentrations near the 903 Area because we excluded larger particles (>30  $\mu m$  AED) from the 903 Area source term, as discussed previously. Most of the other inventory estimates overlap the uncertainty bounds of our inventory estimate. Only Krey (1976) shows a substantially higher inventory estimate than the other researchers.

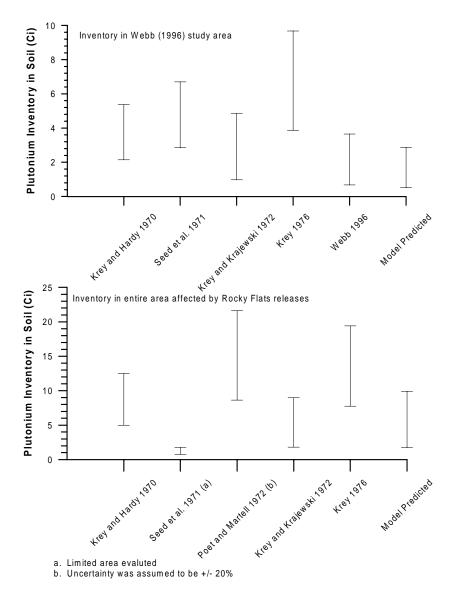
Plutonium inventories in the entire model domain (lower graph in Figure 14) show our estimate was generally in good agreement with the other estimates. The plutonium inventory estimate made by Poet and Martell (1972) appears to be out of line with other estimates. Poet and Martell (1972) used the measured ratio of <sup>239</sup>Pu to <sup>90</sup>Sr in soil samples collected 48 to 65 km (30 to 40 mi) northeast of the RFP to determine the fraction of the plutonium measured in that region attributable to RFP sources. This technique has some inherent problems given the different environmental mobilities of the two radionuclides over time, and it could lead to an estimate that does not reflect what is actually present.

The RFP inventory estimate made by <u>Seed et al.</u> (1971) (lower graph in Figure 14) was for a very restricted area only, and it is probably not representative of the total RFP plutonium inventory. The Seed et al. inventory estimate for the Webb study area is comparable to other estimates (lower graph in Figure 14) when the same area is considered.

Overall, the model-estimated plutonium inventory in soil is within about a factor of 2 of the independent inventory estimates based on plutonium soil concentration data. These results provide some validation of the total quantities of plutonium released from the plant. Most of the plutonium in soil in the model domain is estimated to have originated from the 903 Area.

Contributions to the total plutonium soil inventory in the model domain from the 1957 fire event were between 0.44 Ci (5<sup>th</sup> percentile) and 2.6 Ci (95<sup>th</sup> percentile) based on model simulations. Estimated soil concentrations from routine operations and the 1969 fire would be difficult to discern from fallout plutonium soil concentrations. Far less plutonium was estimated to be deposited in the model domain from the 1957 fire than from 903 Area releases. We attribute

this observation to (a) particles sizes for the 1957 releases were smaller than 903 Area releases, resulting in lower gravitational settling velocities and deposition, (b) winds were relatively light during the 1957 fire compared to the discrete 903 Area releases and resulted in lower calculated deposition velocities, and (c) 1957 fire releases were from an elevated plume compared to ground-level releases for 903 Area.



**Figure 14.** Comparison of plutonium soil inventory estimates from soil concentration measurements. Model Predicted shows the estimates made in this study. Inventory estimates for Webb's study area (209 km² [81 mi²]) are shown in the upper graph. The lower graph represents the entire area affected by all the RFP releases. Inventory estimates were obtained from Table 2-2 in Webb (1996). The 5<sup>th</sup> and 95<sup>th</sup> percentiles of the inventory estimate distributions are shown.

# **Comparisons with Air Measurements**

Comparisons with annual average ambient air measurements were hampered by lack of quality data before 1970. Consequently, comparisons were performed for post-1970 data only. Comparisons in this medium tested the resuspension portion of the model because releases after 1970 were dominated, for the most part, by resuspension of contaminated soil from the 903 Area rather than routine releases from the plant operations. Estimated concentrations onsite and at the new RFP boundary were generally within the range of measured values, with the exception of several years. The model underestimated concentrations at community locations in the late 1970s and early 1980s. However, many of these measurements were below the Atomic Energy Agency's minimum detectable concentration (MDC) and some measurements were less than fallout concentrations in Denver. Negative model bias (model underestimation) during the 1970s and 1980s would not substantially underestimate inhalation exposure for persons who lived in the model domain before this time because exposures were considerably less for those two decades than in the 1950s and 1960s.

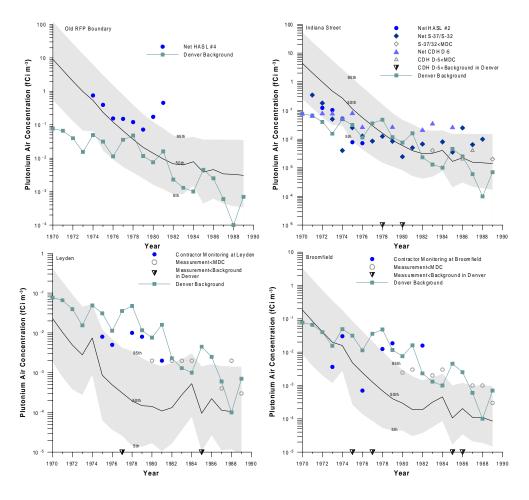
Four monitoring stations were selected for evaluating the annual average plutonium concentrations in air, and they include measurements made by the RFP contractor, the Atomic Energy Commission Health and Safety Laboratory (HASL), and the CDPHE. The four locations were (1) the old RFP boundary where it intersects with the east access road, (2) Indiana Street where it intersects with the east access road, (3) the City of Broomfield, and (4) Leyden. Samplers maintained by HASL were located at the old RFP boundary (HASL 4) and near the intersection of Indiana Street and the east access road (HASL 2). The RFP contractor stations included samplers S-32/37g located near the HASL 2 sampler and stations in Broomfield and Leyden. The CDPHE sampler D-5 was also located on Indiana Street, but it was located about 500 m (1640 ft) south from the HASL and contractor stations. Annual average measured concentrations at each location were determined by Rope et al. (1999, Table B-14). Annual contributions from fallout sources were estimated from U.S. Public Health Service monitoring of air in Denver and were subtracted from the annual average measured concentrations to yield net annual average plutonium concentrations in air resulting from RFP releases. The net plutonium air concentrations were compared to model estimated air concentrations from the RFP releases (Figure 15).

Estimated concentrations during the 1970s are dominated by resuspension sources, with some perturbations during routine releases from buildings and stacks. The peaks observed in the estimated concentrations in 1974 and 1984 (most pronounced at the Leyden location) reflect routine releases from the Building 771 stack and Buildings 776/777 roof vents. The decrease in the estimated concentrations of plutonium in air from 1970 to 1980 is in response to the decrease in resuspension over time and to a lesser extent, the weathering of plutonium from the 0–3-cm (0–1.2-in.) soil layer. The 903 Area was paved in 1969; therefore, the primary suspension source of plutonium-contaminated soil was eliminated in 1970.

In general, model estimates encompass the measured values until about 1976. After 1976, the model tends to underestimate air concentrations at all locations except Indiana Street. At the Indiana Street location, the model underestimates some of the measurements during the 1981 to 1988 time frame. There is a noticeable increase in the measured concentration during the years

<sup>&</sup>lt;sup>g</sup> S-32 was the sampler designation before 1975. After 1975, the sampler was designated as S-37.

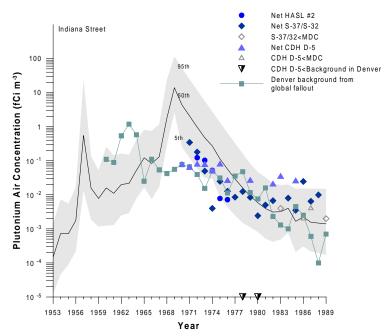
1976 to ~1982 at all stations except Indiana Street. The reason for this increase was not determined. During the 1976–1982 time frame, several of the annual average concentrations at Leyden and Broomfield were either less than Denver fallout or less than the Atomic Energy Agency's MDC. These measurements should be interpreted with caution because of the large uncertainty associated with them. After 1983, all annual average concentrations at the Leyden and Broomfield stations were less than the RFP contractor's MDC. Samplers monitored by the RFP contractor and CDPHE at Indiana Street during the 1982–1989 time frame had several measurements less than the MDC.



**Figure 15.** Comparison of estimated annual average plutonium concentrations in ambient air with measurements at four locations in the model domain. The 50<sup>th</sup> percentile values are shown as the dark line in the shaded region that represents the 5<sup>th</sup> to 95<sup>th</sup> percentile range. Measured concentrations in Denver are also shown and are consistent with global fallout concentrations in other U.S. cities.

It is interesting to note that the measured concentrations at the old RFP boundary between the years 1974–1979 (HASL 4) fall off with about the same slope as the estimated values. This observation provides some validation of the time-dependent soil resuspension model used in this study. In 1980–1981, measured concentrations at HASL 4 increased substantially probably because of resuspension resulting from vehicular traffic.

Figure 16 shows the estimated annual average concentration of plutonium in air at Indiana Street from 1953–1989, along with measured data taken there after 1970. It demonstrates the overall magnitude of exposure from RFP and fallout sources during the operational period of the RFP (1953–1989). The peak annual average concentrations of plutonium in air were about 1 to 2 orders of magnitude greater than concentrations after 1970, and during some years, fallout concentrations exceeded estimated concentrations from RFP sources.



**Figure 16.** Annual average plutonium concentrations in ambient air from 1953 to 1989 at Indiana Street and plutonium-specific measurements taken from 1970 to 1989. The 50<sup>th</sup> percentile values are shown as the dark line in the shaded region that represents the 5<sup>th</sup> to 95<sup>th</sup> percentile range. Plutonium from global fallout as measured in Denver (shown separately) has been subtracted from the measured values.

# **Comparisons with Measurements in Other Media**

Environmental monitoring data for plutonium concentrations in lake sediments and vegetation were also identified as potentially useful for model validation purposes. However, additional modeling and assumptions were necessary to estimate plutonium concentrations in these media. The model-estimated plutonium concentrations in Standley Lake sediments appeared to be underestimated. This underestimation was attributed to underestimating plutonium deposition in the vicinity of Standley Lake and soil erosion and fluvial processes that were not included in the model. The estimated plutonium inventory in Great Western Reservoir from airborne plumes matched values estimated by Schoep and Whicker (1995) and Thomas and Robertson (1981) reasonably well.

Estimated concentrations in vegetation generally matched the temporal trends observed in vegetation measurements taken between 1953 and 1964. The model used to estimate

concentrations in vegetation appeared to overestimate concentrations from the 1957 fire. However, a lack of specific information about vegetation sampling methods and uncertainty in the vegetation model itself made model comparisons in this medium somewhat tenuous.

### PUBLIC INTERACTION AND RISK COMMUNICATION

Interest and questions from the public have played a significant role in the scope and direction of the project. Throughout the course of Phase II, the HAP and RAC have maintained an open dialogue with members of the public and have tried to respond to questions and ideas to the greatest extent possible. We have communicated results of our ongoing work regularly to solicit input on our approaches to various aspects of the project, to request additional sources of information, and to inform stakeholders about the progress of the work. It was recognized early on that public concerns would be very important for both the credibility of the research and for identifying issues that were perceived to be critical. As a result, a number of changes were made in the direction of the research to help respond to questions raised.

Several examples of questions raised by the public that were addressed during the study follow.

- Concern about protecting the confidentiality of workers who were interviewed for the project led the *RAC* and the HAP to develop a confidentiality policy in September 1994.
- Questions about the document review process, especially the review of classified records, led to developing simple document databases that were available to the HAP and to the public upon request. The database tracked the classified records and monitoring records that we reviewed during Phase II. This concern also prompted a citizen member of the HAP to apply for and obtain a clearance from DOE to see classified material. This member then accompanied us to the site to help review classified records.
- Because of concern about the quality and analysis of historical measurements and data reported by the RFP operator, RAC expanded the data quality section in their project report dealing with the analysis of environmental monitoring data (Rope et al. 1999). To supplement these historic records, other options were continually pursued to verify releases to the environment. For example, historic aerial photographs and government satellite imagery were used in an attempt to visually check for evidence of plumes from some of the fires at the site.
- A mass balance approach of the onsite processes was carefully evaluated to estimate the amount of contaminants released to the environment from the RFP as a way to set an upper bound limit on some of the releases from the site.
- Concern about selecting important contaminants of concern during Phase I of the project led to a careful reexamination of chemicals used or produced at the RFP. Some of the contaminants of concern that were reevaluated during Phase II were dioxin, methyl ethyl ketone, ethylene oxide, propylene oxide, asbestos, uranium, and tritium releases to surface water. We also verified the location of the Ralston School in the late 1950s. This was an important monitoring location following the 1957 fire.

Early in Phase II, we met with members of the public to identify and discuss their concerns about the project. Transcripts of this meeting were recorded and used to understand criticisms about the project up to that point. These issues were catalogued and listed in a notebook for a

permanent project record so each one could be addressed to the best of our ability. As Phase II continued, *RAC* added to the list of citizens' concerns and developed responses for them. It was not possible to respond to every issue, but from 210 questions and issues that were raised, responses were developed for more than 200 of them. The few that remain unresolved are issues that lie being beyond the scope of the study or that lack historical information with which to work. Three issues that could not be resolved are listed here.

- 1. It is important to better understand the significance of sources of uranium, particularly depleted uranium, which were used at the RFP site. This item has been listed as one needing further research. Nevertheless, it is not likely that the use and release of depleted uranium at the site would significantly affect the risk estimates, rather it is more a question of thoroughness of source term evaluation for this radioactive material.
- 2. The final disposition of the waste drums from the major fire events and reports that some were stored in Building 991 could not be verified. This issue is difficult to completely address because of the lack of knowledge about the number and contents of waste drums shipped offsite.
- 3. Treating entire contaminated buildings onsite as release points, or source terms, may be an important issue as demolition of buildings and cleanup at the site moves forward. However, this issue is outside the scope of the current project that looked at past releases.

Throughout the project, *RAC* has responded to these questions, concerns, and comments from the public and those closely involved in the process. We compiled these questions and concerns taken from letters, written reports, electronic mail, and questions at public meetings. Questions raised by the public and responses to the questions have been compiled and are available in a project report (Meyer et al. 1999).

Meetings of the HAP were open to the public and evening public meetings were also held to report progress. During the course of Phase II, public meetings were held on a variety of topics from atmospheric transport to developing source terms for releases to the environment. These meetings and one-on-one conversations with interested members of the public were a primary means of communicating the technical work as it progressed and our findings. In addition to direct communication with people, newsletters, fact sheets, and less technical summaries of technical reports were also employed.

Dose reconstruction relies not only on historical records but also on information, either oral or written, given to researchers by people with firsthand knowledge and scientists, workers, and members of the public who have researched or obtained information that may be useful in the study. Because of the sensitive (not classified) nature of some information, there is often a need to protect the identity of individuals. Therefore, a policy was developed to provide guidance on interviews conducted as part of the Historical Public Exposures Studies on Rocky Flats. This policy outlined specific steps that would be taken by the HAP to ensure the confidentiality of individuals who wish to provide information that may be applied in the dose reconstruction for the RFP. Although the policy was not used extensively, we did explain it to people being interviewed in case they wanted to remain anonymous. Development and approval of this policy by the HAP and *RAC* was an important contribution to the study that could be used in future projects.

As with any public study, inquiries from people interested in the research are a vital component of the project's success. We believe that extraordinary efforts were made during the

Historical Public Exposures Studies on Rocky Flats to listen carefully to questions being raised and to respond to these questions to the greatest extent possible. Recommendations and questions from the public played a major role in the scope and outcome of the project.

### CONCLUSIONS AND RECOMMENDATIONS

The results of the Historical Public Exposures Studies on Rocky Flats show that the primary risks posed to the public from operations at the RFP were from plutonium and carbon tetrachloride. These two materials were determined to be the most important contributors to cancer incidence risk from approximately 8000 radioactive and nonradioactive materials that were present at the plant during operations. Inhalation was the most important exposure pathway for these materials. For plutonium, cancer of lung, liver, bone surfaces, and bone marrow (leukemia) are the endpoints (or health effects) of concern and account for ~97% of the total risk. For carbon tetrachloride, liver cancer is the endpoint of concern. Other pathways were much less important, such as ingestion of food and water that contained plutonium or carbon tetrachloride and external exposure from radioactivity in air or on the ground. *RAC*'s estimates of risk were based on a number of scenarios that characterized the lifestyle, period of residence, gender, and age of hypothetical individuals in the Rocky Flats area.

The major event contributing the highest individual risk from plutonium released from Rocky Flats was the 1957 fire. This finding is important and is different from previous analyses of risk at the site that had shown the 903 Area was the major contributor to risk historically. However, exposure and risk from the 1957 fire were contingent on being located in the path of the plume during the small number of hours that the release occurred. Near ground level, the plume from the fire traveled southward, toward Arvada, then northeast toward Northglenn. The amounts of material deposited to soil from the 1957 fire were small because predominantly small particle sizes were released. This makes it difficult to verify the direction of the plume during the fire using soil sampling data. In addition, detailed offsite detailed soil sampling was not performed until after the major 903 Area releases. Releases from the 903 Area during 1964–1969 were the second largest source of risk from plutonium. Approximately 90% of the releases occurred on 6 days during that time period. A wide range of particle sizes was released from the 903 Area and was responsible for most of the measurable offsite contamination from RFP sources.

Of the scenarios that were modeled, a laborer living and working southeast of Leyden near Indiana Street and  $64^{th}$  Avenue throughout the entire period of operations at the RFP has the highest risk of  $(50^{th}$  percentile) developing cancer from plutonium inhalation. These risks range between  $2.0 \times 10^{-4}$  (95<sup>th</sup> percentile) and  $2.2 \times 10^{-8}$  (5<sup>th</sup> percentile), with a median risk estimate of  $2.3 \times 10^{-6}$ .

The risks of cancer incidence from carbon tetrachloride releases were estimated to be comparable to those from plutonium. However, unlike plutonium, carbon tetrachloride has not been demonstrated to cause cancer in humans. The EPA classifies carbon tetrachloride as a probable human carcinogen. Also, few data were available to verify the volumes of carbon tetrachloride released; therefore, the risk estimates for carbon tetrachloride are inherently more uncertain than those for plutonium.

These studies represent a major contribution to the understanding of historical operations and releases from the Rocky Flats facility since it opened in 1952. During the course of the work,

RAC made every effort to find and obtain historical records that could provide information useful to the research. During this process, many historical records were declassified so they could be publicly available to researchers. As with any dose reconstruction covering a large period of years, some data that would have been helpful could not be located. However, it was possible to make defensible assumptions about missing data to fill gaps using a number of techniques that have evolved during the recent decade of dose reconstructions on DOE sites. In addition, considerable effort was made during the study to use environmental monitoring data collected over the years from a number of different sources to help confirm RAC's estimates of releases and estimated transport through the environment. The results of these validation efforts provided confidence in the results.

There are several important contributions to science and public policy from the Historical Public Exposures Studies on Rocky Flats that are noteworthy. First, and most importantly, these results help people who lived near the RFP understand what contaminants they may have been exposed to and the associated risks. It is important that everyone who lived near Rocky Flats has access to this knowledge. It is also important to have a thorough understanding of the past because the site is no longer operating and parts of it will eventually be released for public use after clean up and remediation activities have been completed.

The study provided a number of new contributions to the development of technical methods that will be of value in future risk assessments. Examples of these contributions include (a) reconstructing the releases of plutonium to the atmosphere (source terms) from the 1957 fire, (b) evaluating resuspension of particles during high wind events that occur in the Rocky Flats area, (c) comparing five air dispersion models to select the model most appropriate for the analyses considering the data available and the geographic location, and (d) estimating organ-specific cancer risk following inhalation of plutonium.

The source term derived for the 1957 fire was unique because of the "inside out" approach that was used to generate it. The Phase I estimate of releases of plutonium from the fire was based primarily on measurements of plutonium in the environment. *RAC*'s estimate of the source term employed a new technique that had not previously been used for reconstructing releases from the 1957 fire at the RFP. The Phase II release estimates from the fire were considerably higher than Phase I.

The study investigated suspension of plutonium-contaminated soil from the 903 Area during high wind events. During the course of this work, *RAC* uncovered previously unused meteorological data that were suitable for atmospheric transport modeling and calculation of wind-driven suspension from the 903 Area. Efforts during Phase I failed to identify this information and, therefore, the previous 903 Area releases were evaluated as a steady-state process over a 5-year period (1964–1969).

Median estimates of respirable release quantities estimated in Phases I and II differed by, at most, approximately a factor of 2 (0.34 Ci in Phase I compared to 0.6 Ci in Phase II). Maximum concentrations estimated during the Phase I study were 2 to 3 times higher for Phase I compared to Phase II estimates. These concentrations differed because the Phase I assessment assumed that the activity was released over a longer period of time at average wind speeds and the Phase II assessment showed that the majority of releases from the 903 Area occurred during high wind events. High winds result in greater soil suspension but also greater dilution of airborne plumes at downwind locations. Consequently, it appears that while discrete high wind events may have contributed to most of the releases and offsite contamination, they did not lead to proportionally

higher offsite air concentrations. Thus, while the 903 Area is suspected to be the greatest contributor to offsite contamination, it was not the greatest contributor to risk because of the effects of high winds when most of the activity was attached to larger, nonrespirable particles.

It was determined in Phase I that the atmospheric transport of materials was the primary pathway by which people were exposed. Therefore, it was decided very early in Phase II that considerable effort had to be put into selecting an air dispersion model that could cover the geographical area of interest and use the meteorological data available in the historical records. Therefore, it was important to find the right model to fit the geography and the data without an unreasonable expenditure of resources. This decision was made by comparing five different atmospheric dispersion models to see which best suited the study objectives. While no one model outperformed the others in all modeling objectives, the overall performance of RATCHET was best for this project.

One contribution of the study to science was the development of organ-specific lifetime cancer incidence risks with uncertainties for plutonium inhalation. Four independent sources of information were used to estimate the risk coefficients with uncertainties: (1) epidemiologic studies of persons exposed to plutonium, (2) epidemiologic studies of persons exposed to less densely ionizing radiation combined with a relative biological effectiveness factor, (3) epidemiologic studies of persons exposed to alpha-emitting radionuclides other than plutonium, and (4) controlled studies of animals exposed to plutonium and other alpha-emitting radionuclides and extrapolated to humans. The intrinsic merit of these sources was assessed and used to weight the risk estimates for each cancer endpoint. The result was a set of lifetime cancer incidence risk estimates for lung, liver, bone, and bone marrow (leukemia) following plutonium inhalation. The impact of gender and age at exposure on the risk estimates was considered. The approach could be expanded to derive risk estimates with uncertainties for other radionuclide intakes.

Public involvement in the historical dose reconstruction was very important to the success of the project. It was a very open process. Comments and ideas were aggressively sought and had a significant impact on the direction of the research. Examples of issues raised by the public that were addressed in the project include evaluating dioxin released as a result of incinerator operation, estimating risk from beryllium released to the atmosphere, assessing risk of tritium released to surface water onsite, and comparing radiation doses from uranium released from the site to those from plutonium. Each question raised or comment provided was documented and responded to in writing. All of this information was retained and published in one of the final reports for the project. This report will provide a reference for the public if they have questions in the future that were answered by scientists during the course of the project.

In conclusion, Phase II of the Rocky Flats Historical Public Exposures Studies yielded many lessons that must be remembered and applied to future projects. New approaches and techniques that were applied during this historical public exposures study be adapted to both retrospective and prospective risk assessment and should be considered in decision-making about cleanup currently ongoing at the site and at other DOE facilities. Most importantly, the public has a comprehensive analysis that can be drawn upon to understand what happened at Rocky Flats in the past and to identify what their risks might have been from releases of materials to the environment during past operations at the site.

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## **APPENDIX A**

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